Microstructural Investigation of Zr Based Alloys Subjected to Electron, Light and Heavy Ion Irradiation

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Introduction

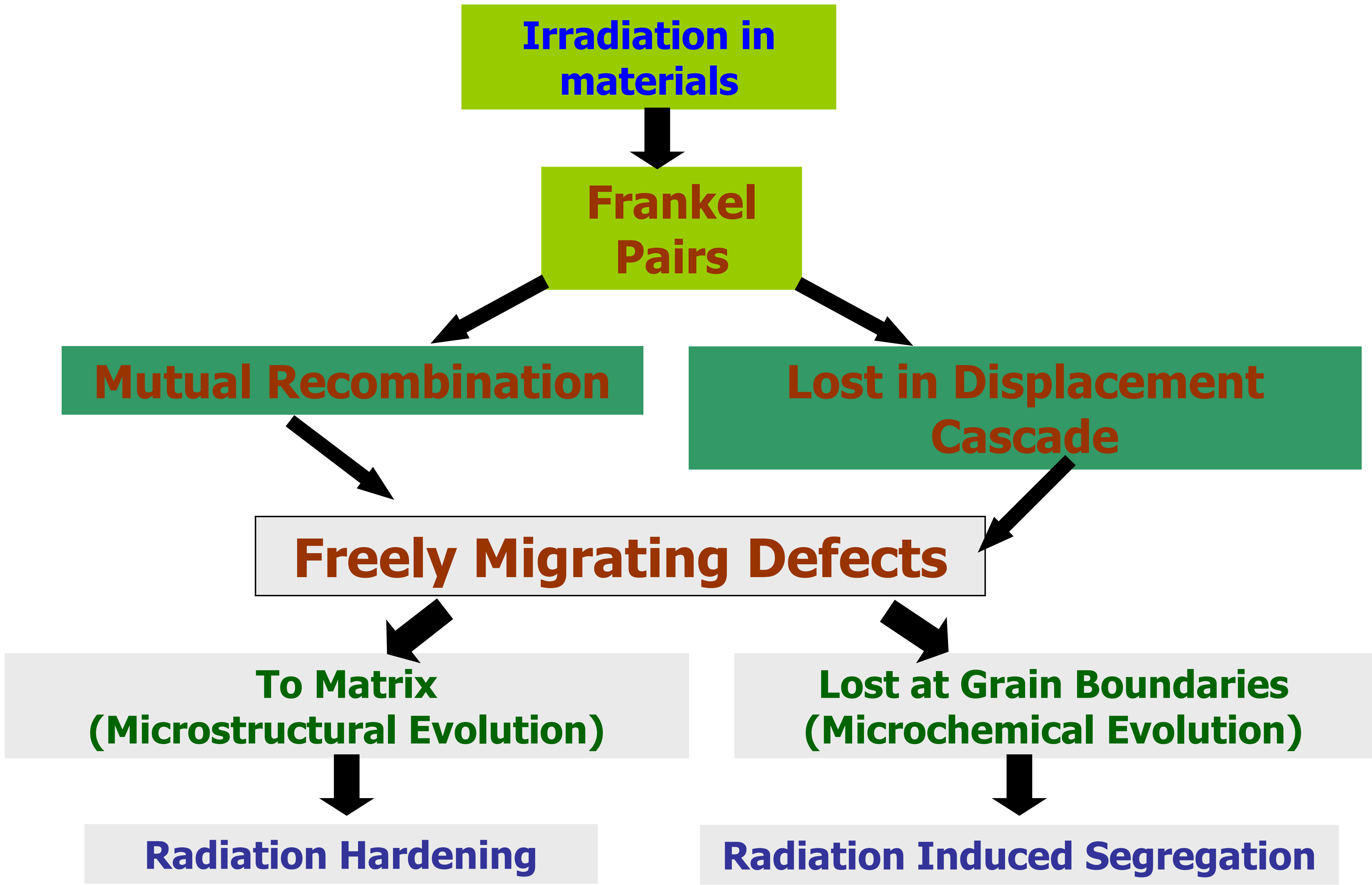
- The irradiating particles impart energy to the target materials by the process of ionization, electronic excitation, displacement of atoms from the lattice, etc.

- Each of these processes of energy transfer is a function of type of ions, energy of the ions and the nature of target materials. The energy transferred to materials is restored in terms of generation of interstitial and vacancy defects, which upon increasing concentration interact in many ways forming deferent kind of defects into the materials.

- In addition to the defects generation, active ions like hydrogen, oxygen appear to participate in phase transformation also.

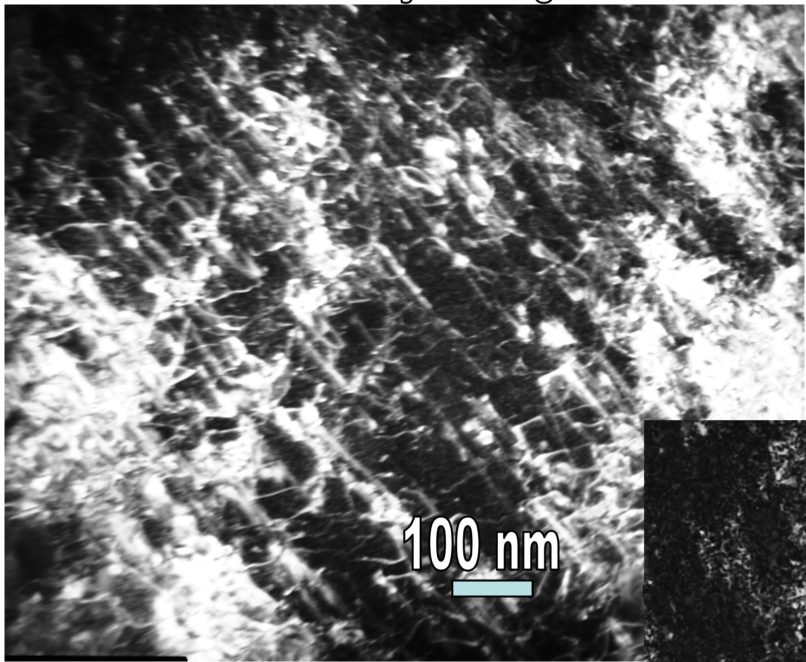
- Irradiation by electrons have also shown to induce phase transformation. Effect of combination of temperature and electron dose is seen in the alloy of Zr-20Nb as initiation of the ω phase. Simulation of SAD patterns show that there was a systematic arrangements of various w unit which lead to the formation of diffuse intensities.

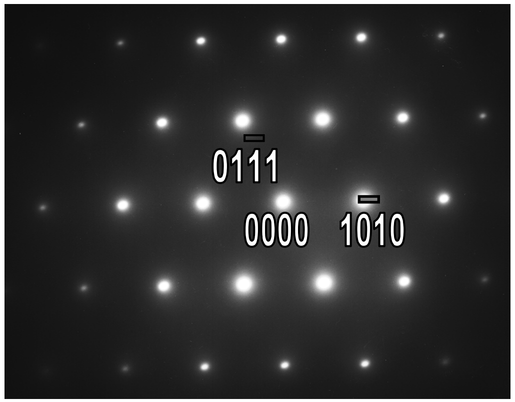
- Irradiation by oxygen showed large amount of defects and tracks of irradiation damages could be seen in the cross section samples. Some of diffraction evidences and energy loss spectra appear to suggest the formation of an oxide phase.

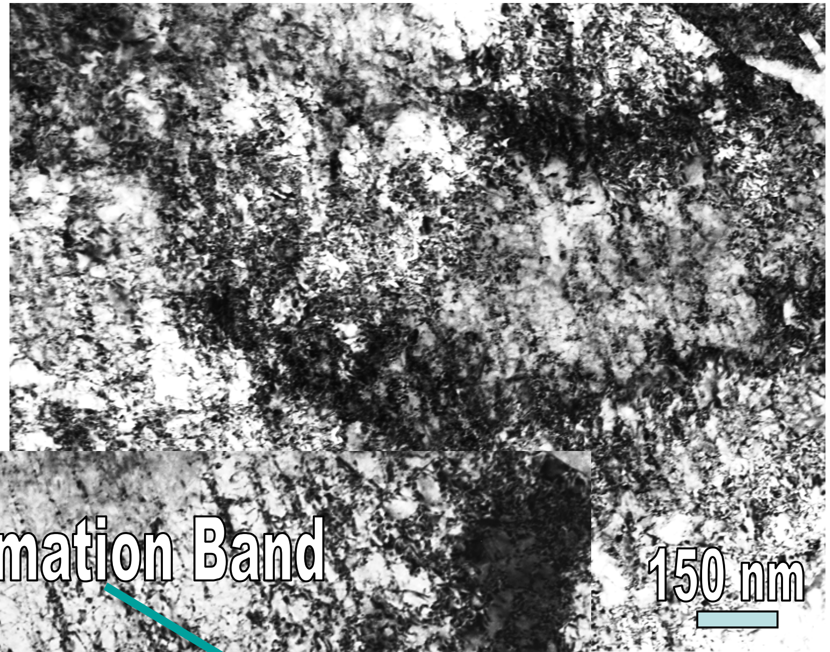

Irradiation in materials
Frankel Pairs
Mutual Recombination
Lost in Displacement Cascade
Freely Migrating Defects
To Matrix (Microstructural Evolution)
Lost at Grain Boundaries (Microchemical Evolution)
Radiation Hardening
Radiation Induced Segregation

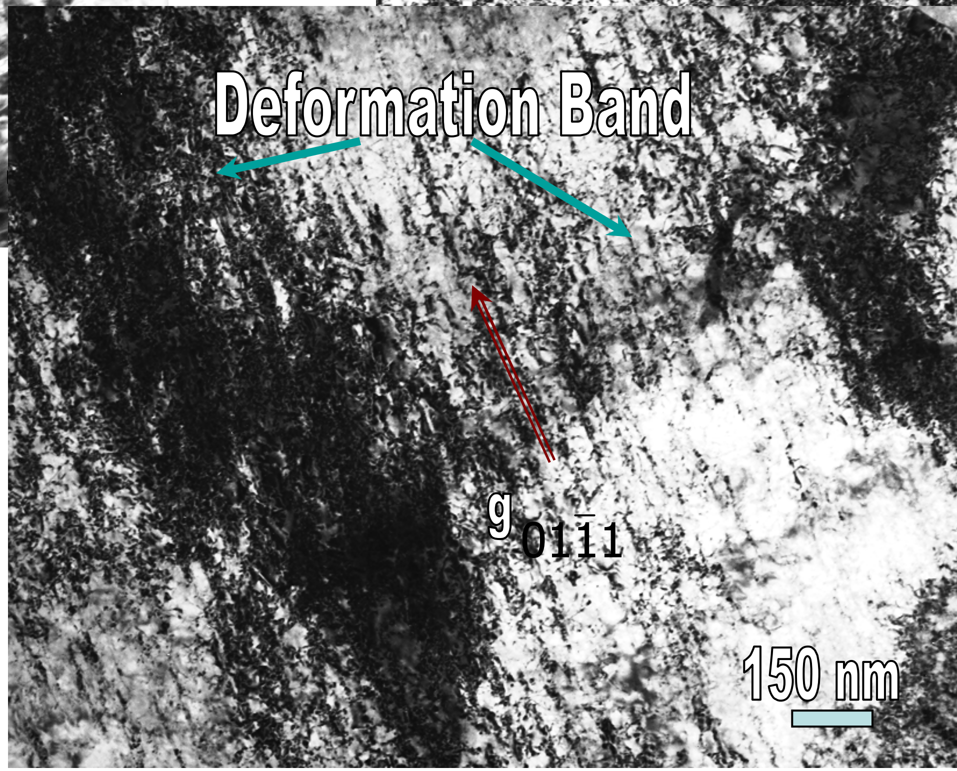
Irradiated Microstructures-formation of defects

Low doses - tendency for alignment



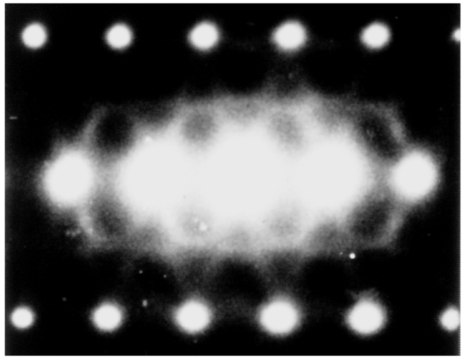


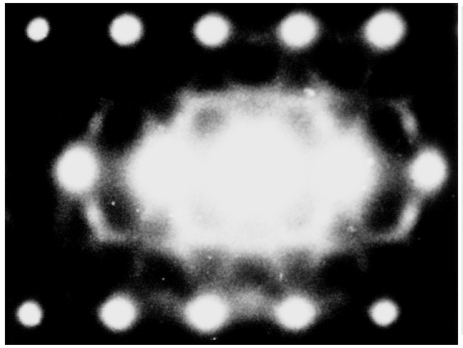


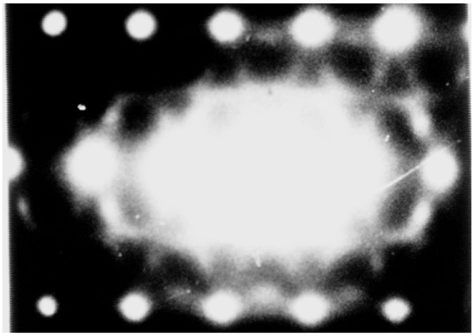


Irradiation Induced ω formation

300 K

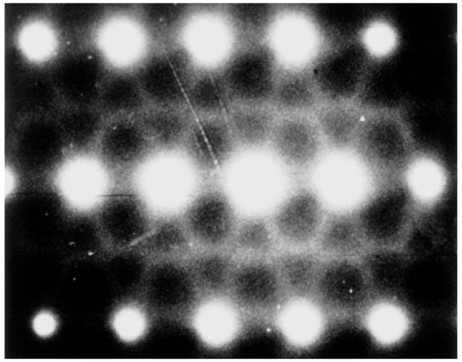


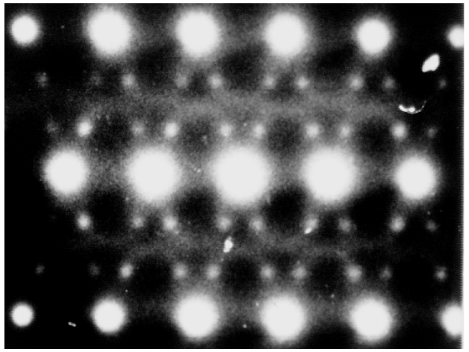


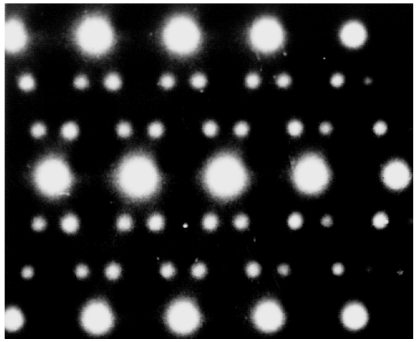


t = 0 s   60 s   300 s

450 K



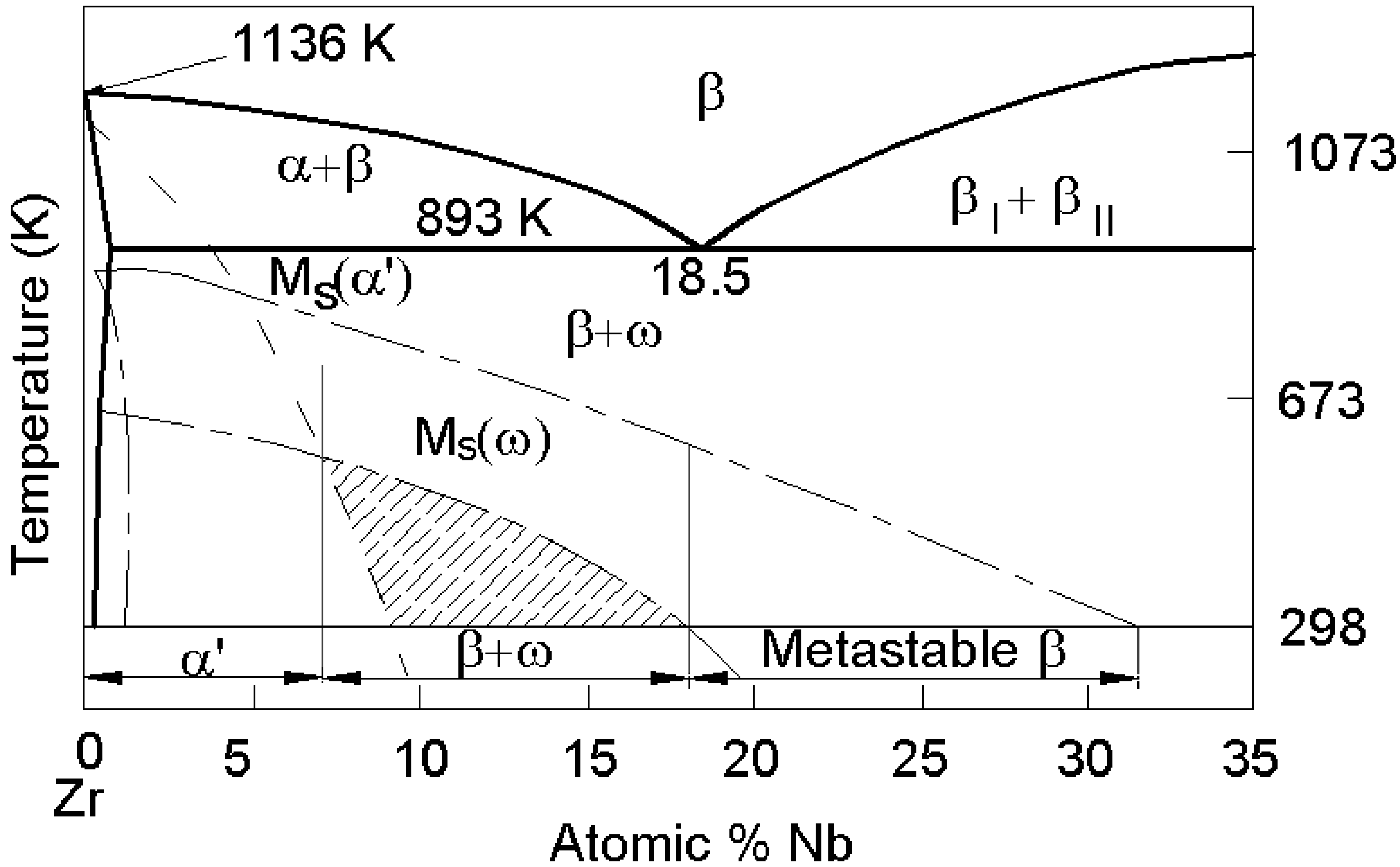




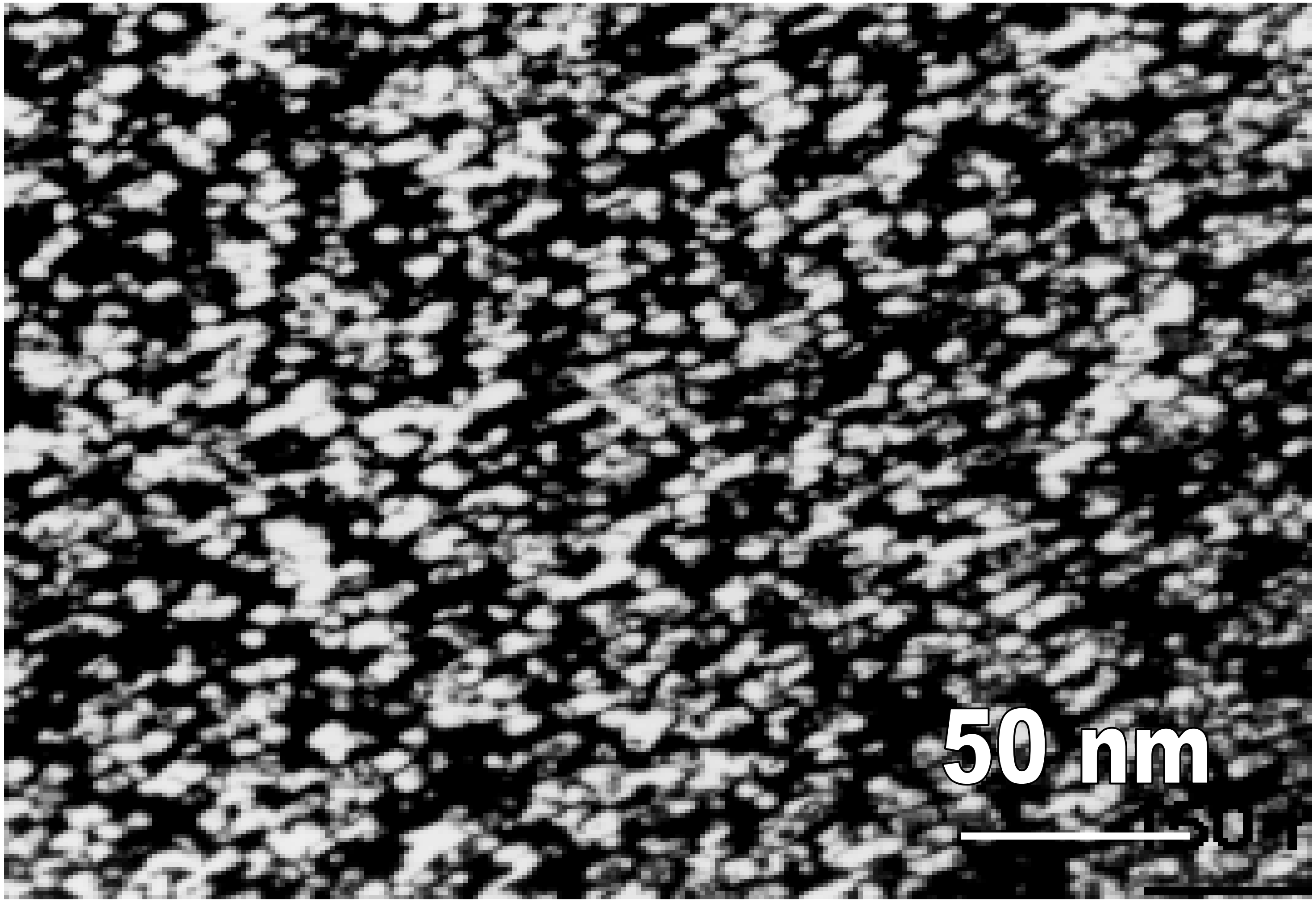
t = 0 s   120 s   240 s

Zr-20Nb , 1 MeV

Binary Zr-Nb phase diagram

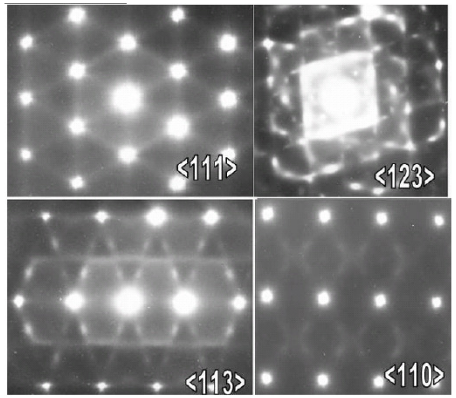


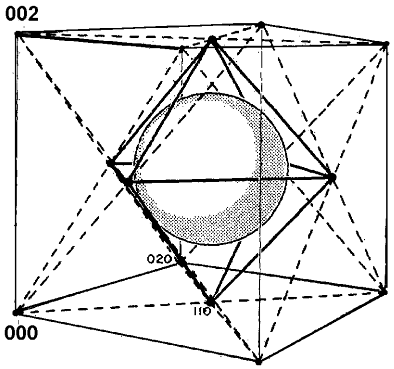
Morphology of thermal ω

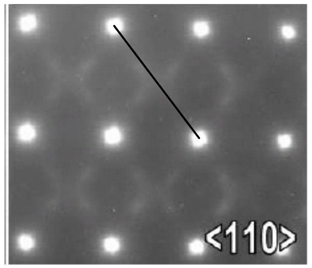


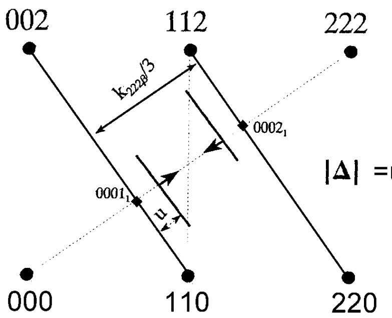
β – quenched-Fine particle ω precipitation

β→ ω phase transformation

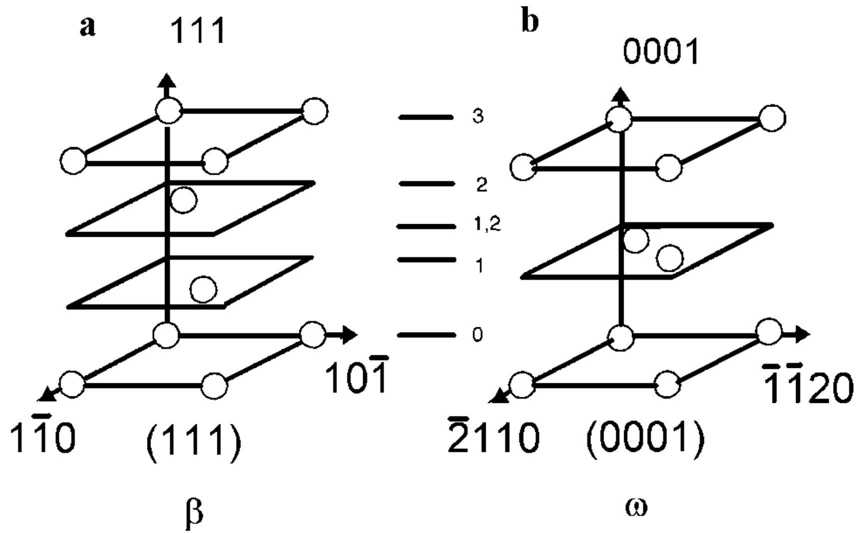


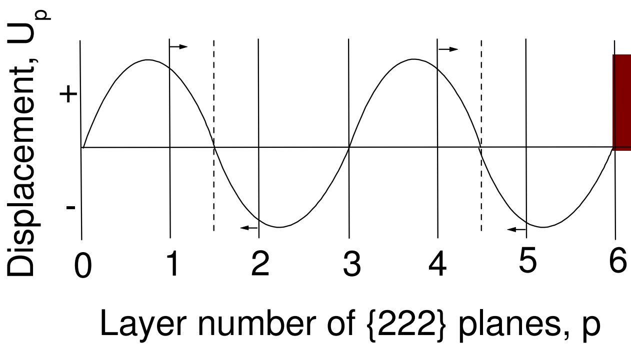




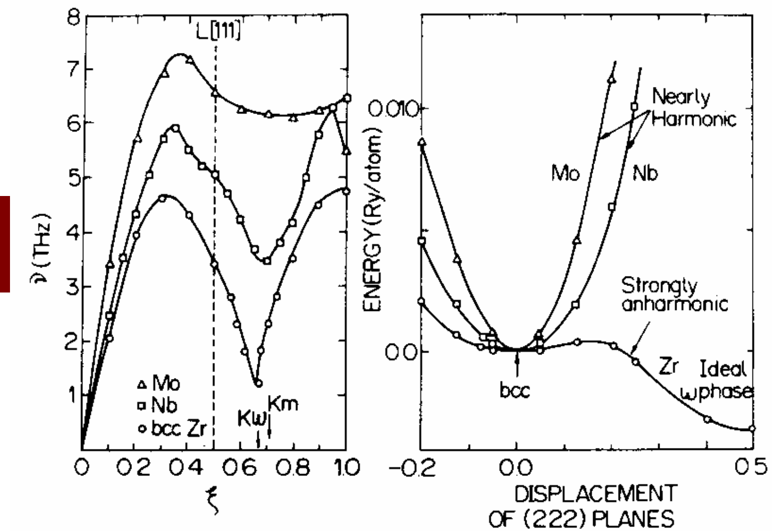


$|\Delta| = u/k_{222_\beta}$

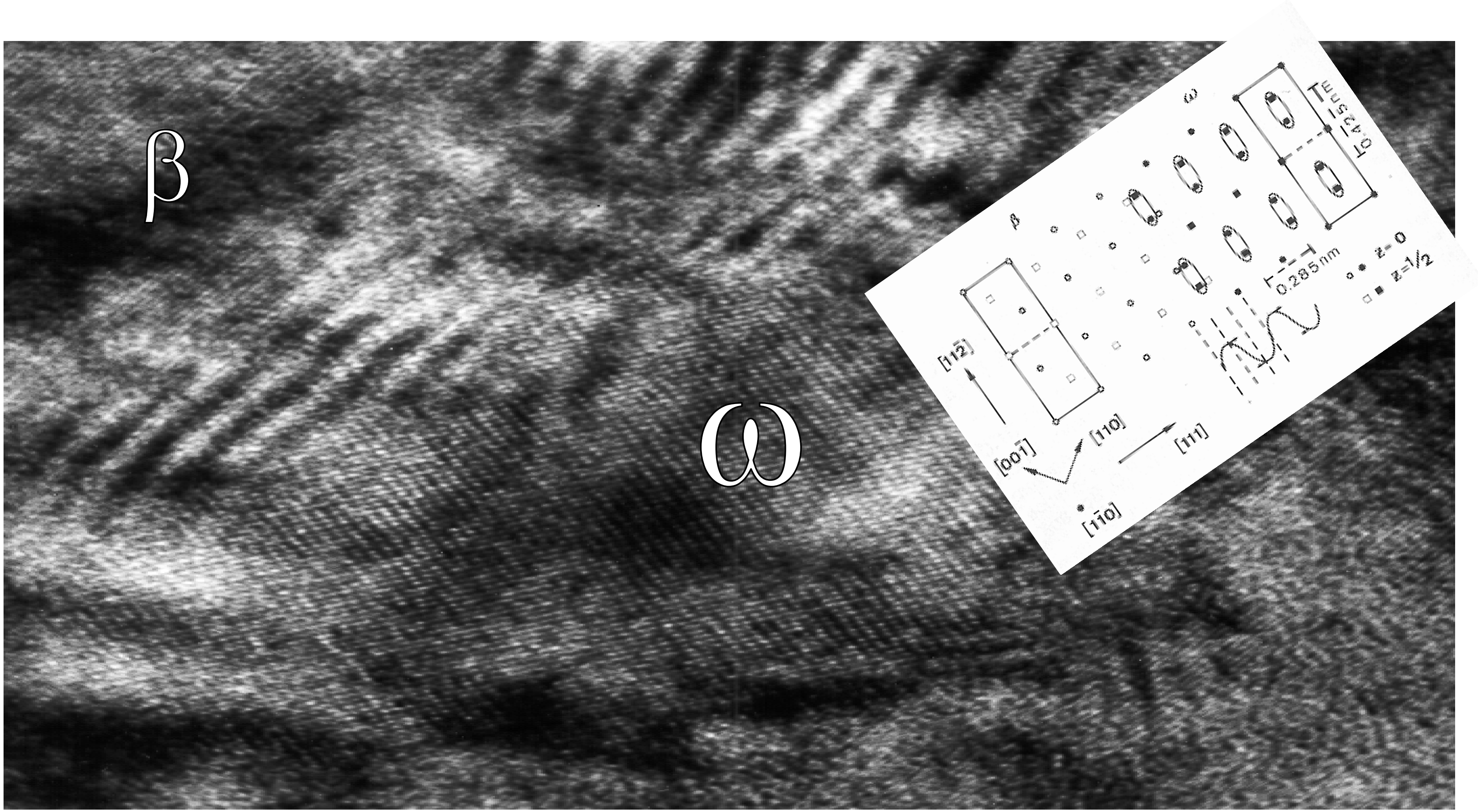


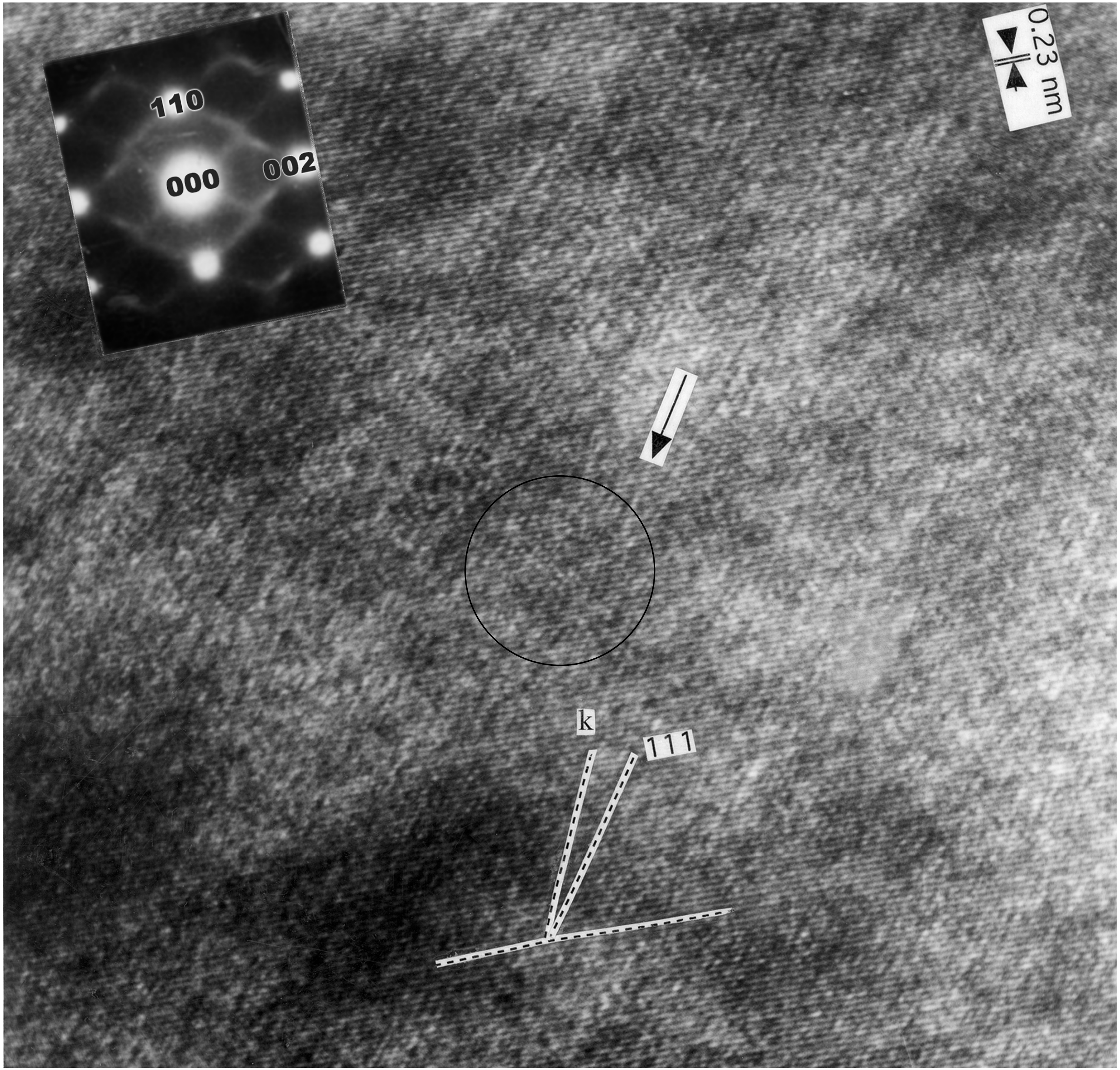


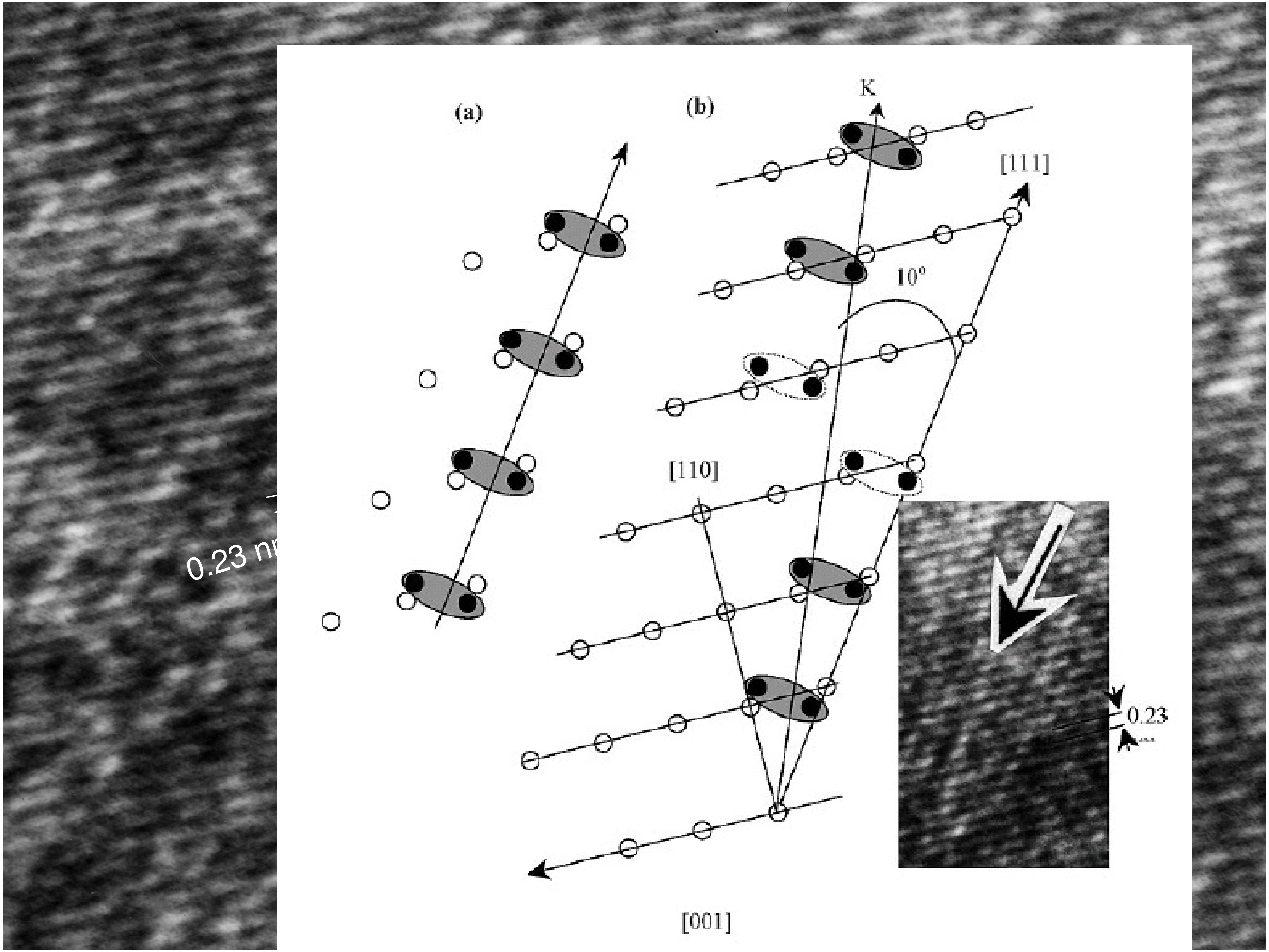
$$U_p = A_d \mathrm{Sin} K_d x_p$$



HREM of image of Aged Zr-Nb alloy

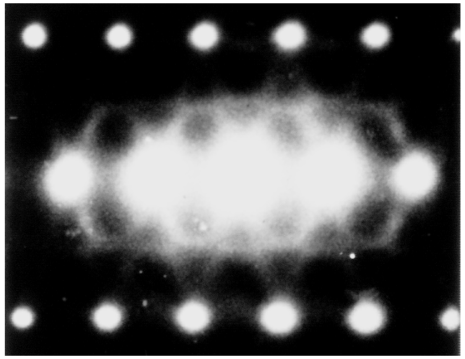



110
000
002
0.23 nm
k
111

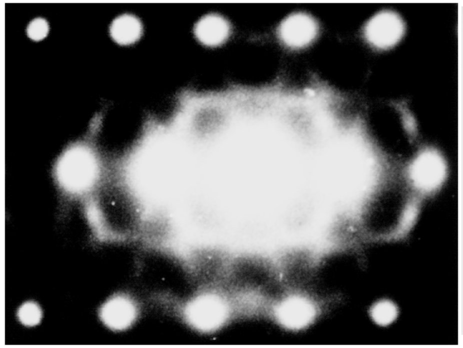

(a)
(b)
K
[111]
10°
[110]
[001]
0.23

Irradiation Induced ω formation

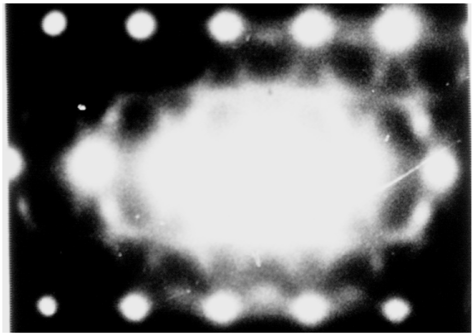
300 K



t = 0 s

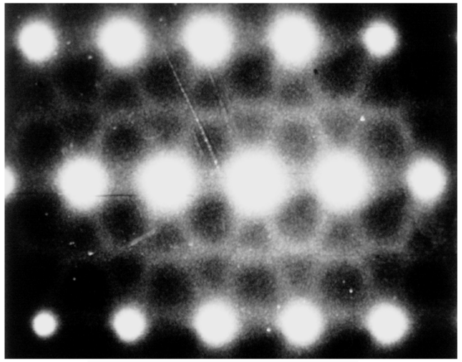


60 s

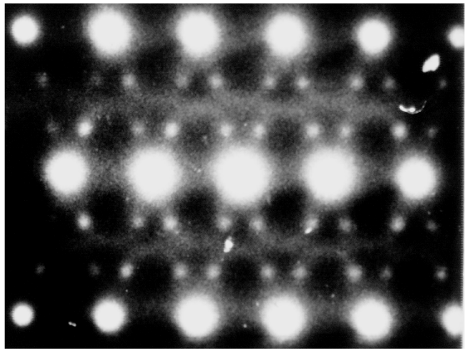


300 s

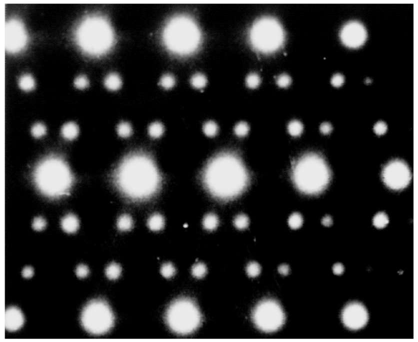
450 K



t = 0 s



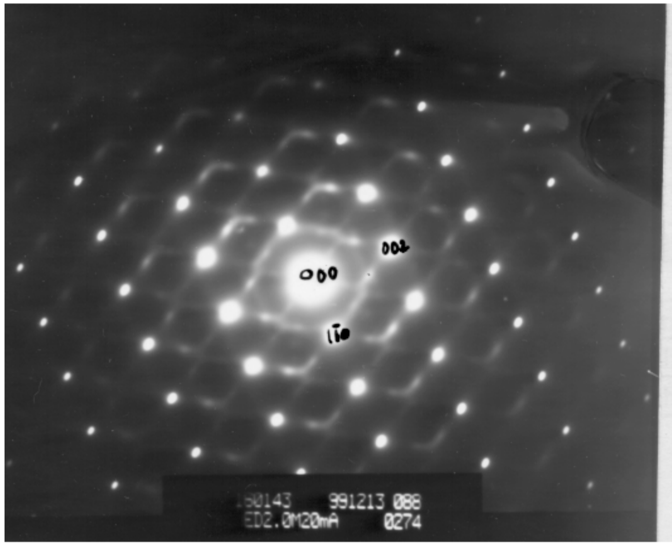
120 s



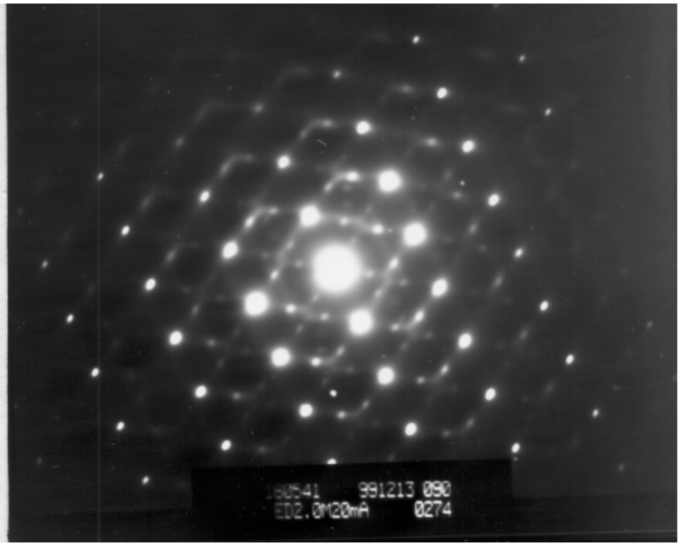
240 s

Zr-20Nb , 1 MeV

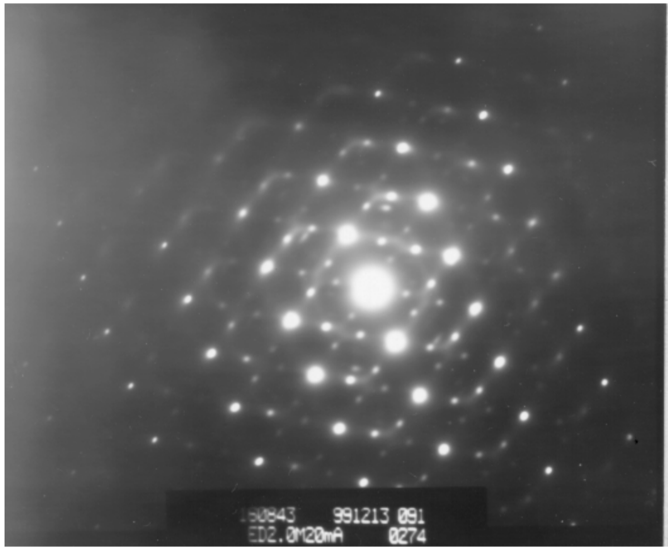
Irradiation Induced ω formation



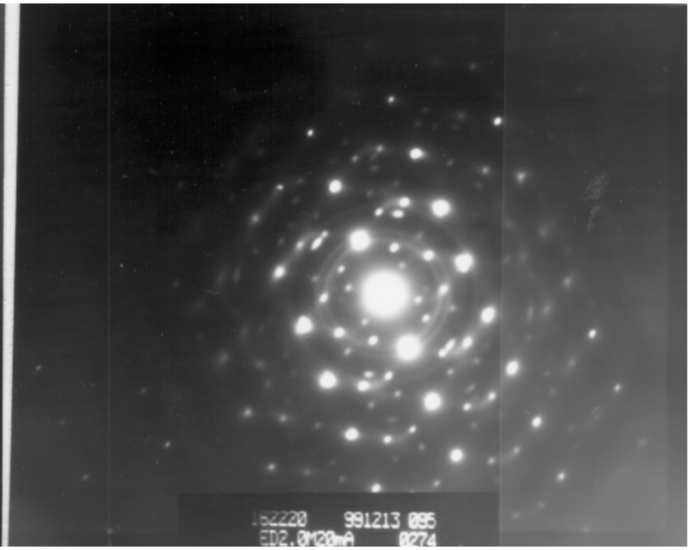
15 sec.



60 sec.



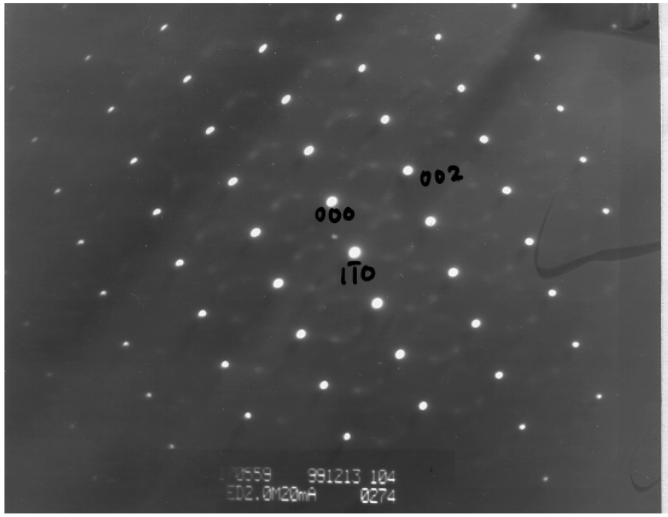
120 sec.



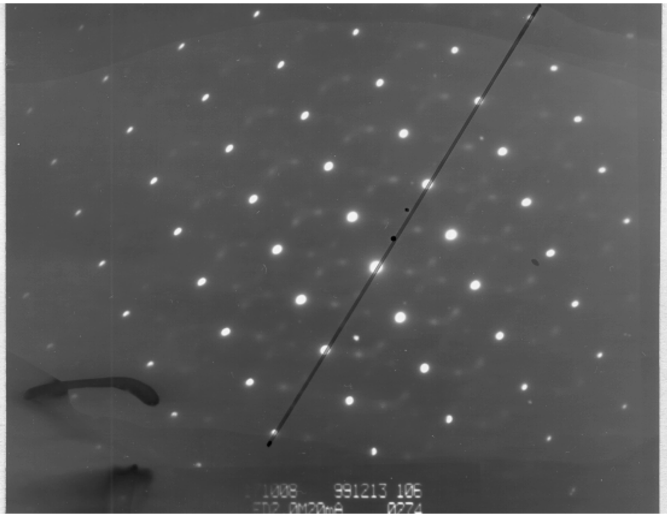
8 min

Zr-20 Nb, T 473 K,
2 MeV e-irradiation

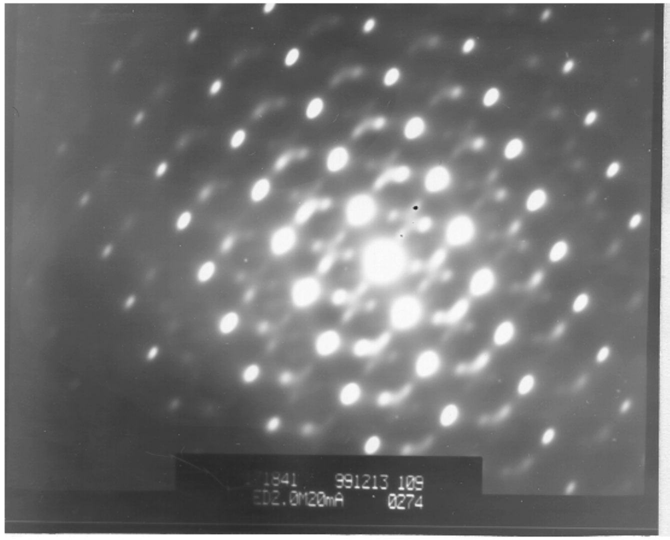
Irradiation Induced ω formation



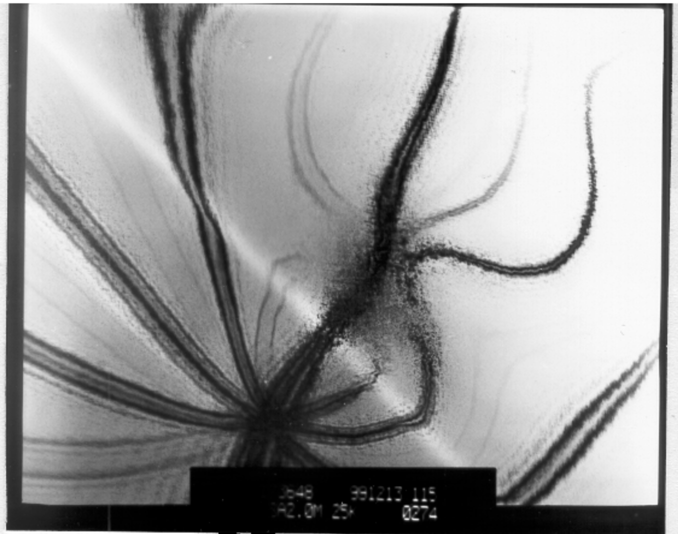
30 SEC.



120 SEC.



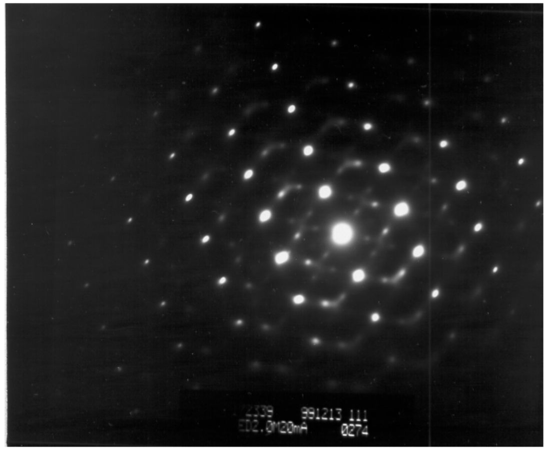
480 SEC



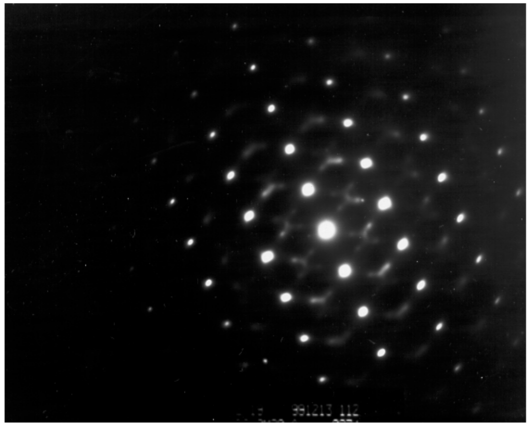
BF

Zr-20 Nb, T 573 K,
2 MeV e-irradiation

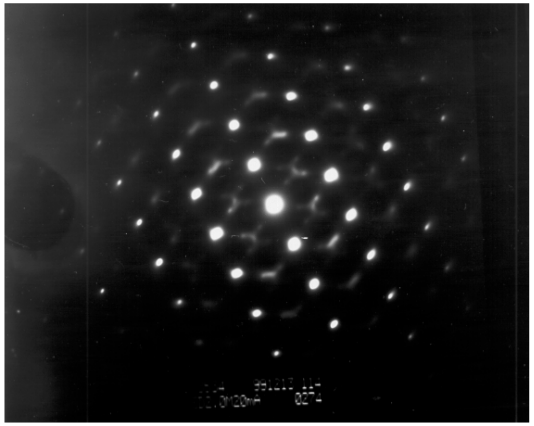
Irradiation Induced ω formation



600 sec



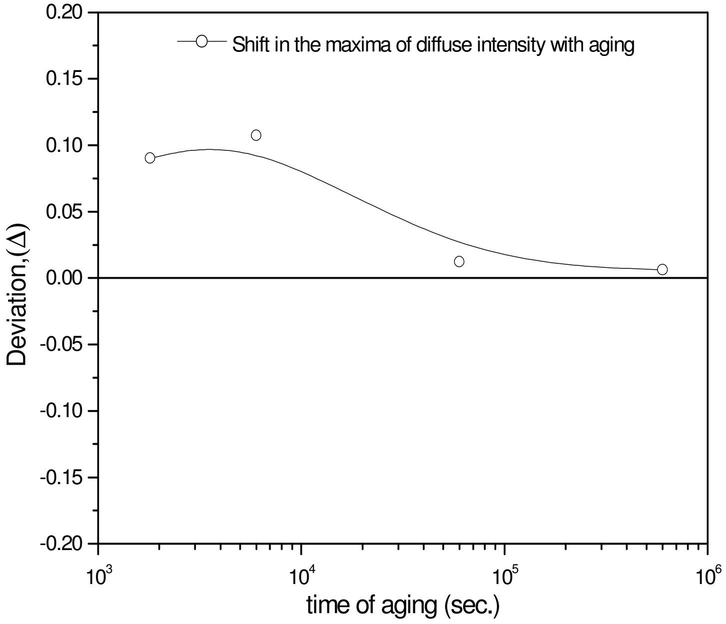
720 sec

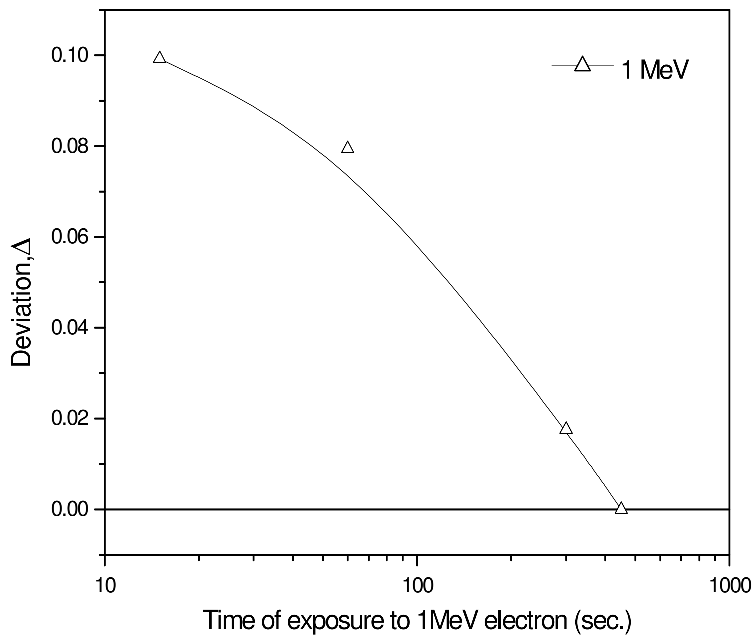


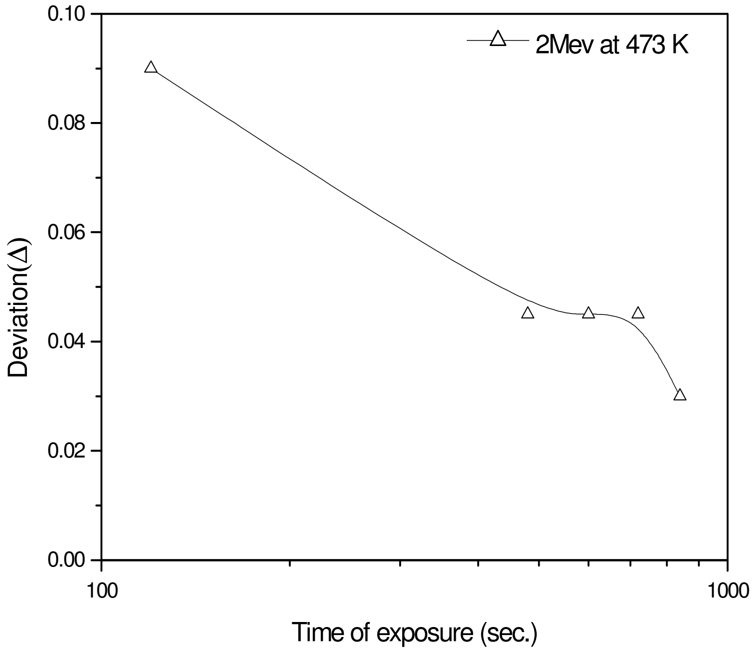
840 sec

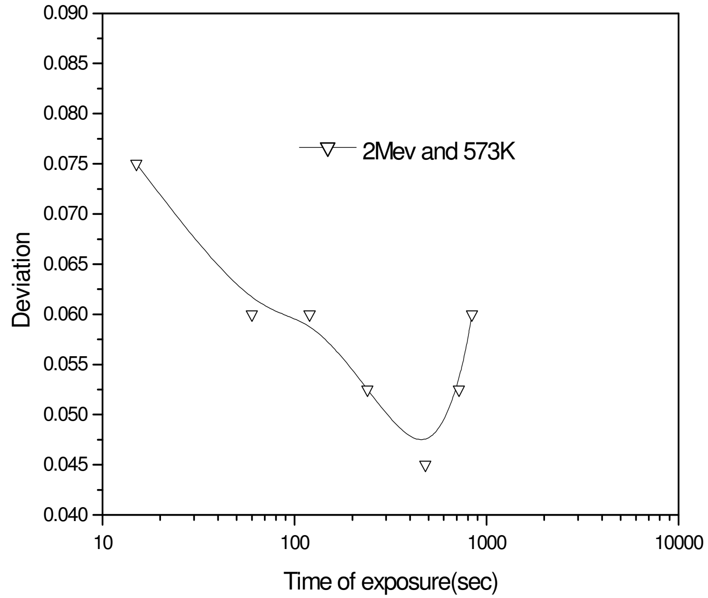
Zr-20 Nb,
T 573 K,
2 MeV e-irradiation

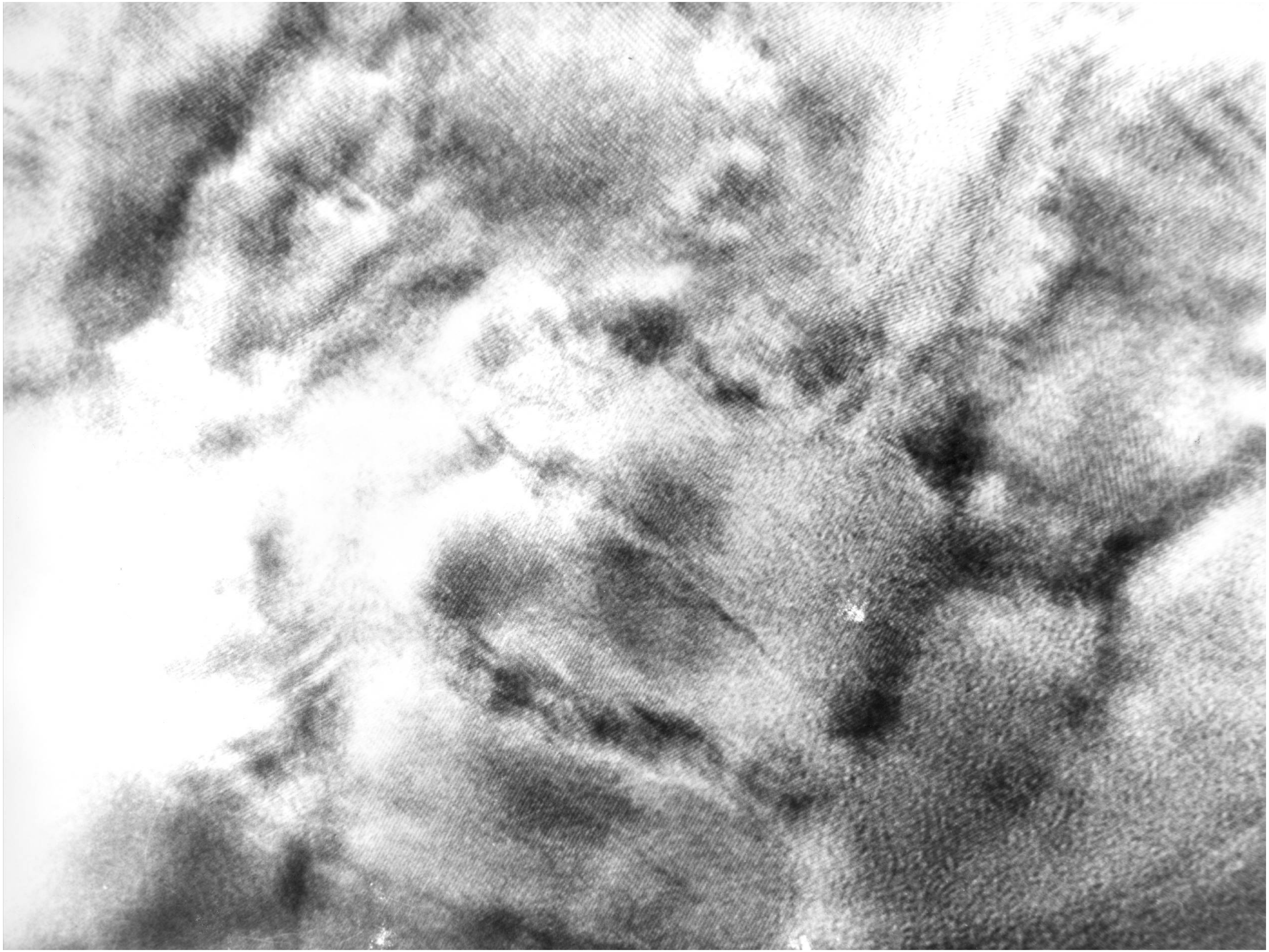
Prolong exposure
initiate dissolution of the ω phase


Shift in the maxima of diffuse intensity with aging
Deviation,(Δ)
0.20
0.15
0.10
0.05
0.00
-0.05
-0.10
-0.15
-0.20
10^3
10^4
10^5
10^6
time of aging (sec.)


1 MeV
Deviation,Δ
0.10
0.08
0.06
0.04
0.02
0.00
10
100
1000
Time of exposure to 1MeV electron (sec.)


2Mev at 473 K
Deviation(Δ)
0.10
0.08
0.06
0.04
0.02
0.00
100
1000
Time of exposure (sec.)


2Mev and 573K
Deviation
0.090
0.085
0.080
0.075
0.070
0.065
0.060
0.055
0.050
0.045
0.040
10
100
1000
10000
Time of exposure(sec)



Simulation of ω precipitation in BCC Zr

Methodology

Create a three dimensional lattice of BCC unit cells (in the present case the lattice is 100X100X100)

Randomly nucleate ω particles (rod shaped particles with length parallel to 111 direction) by proper displacement of atoms.

Calculate the resulting diffraction intensity for appropriate reflections.

Parameters:

The aspect ratio of the ω particle

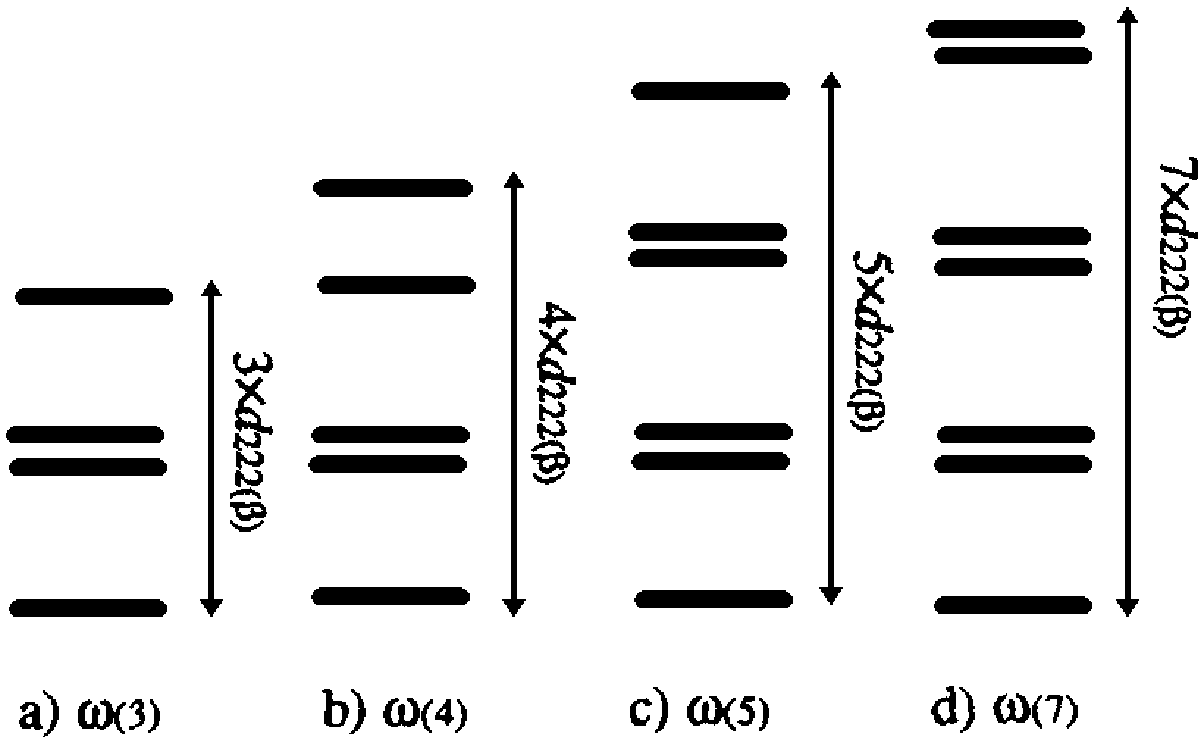
The number of ω rods in the given volume (number density)

Stacking of ω

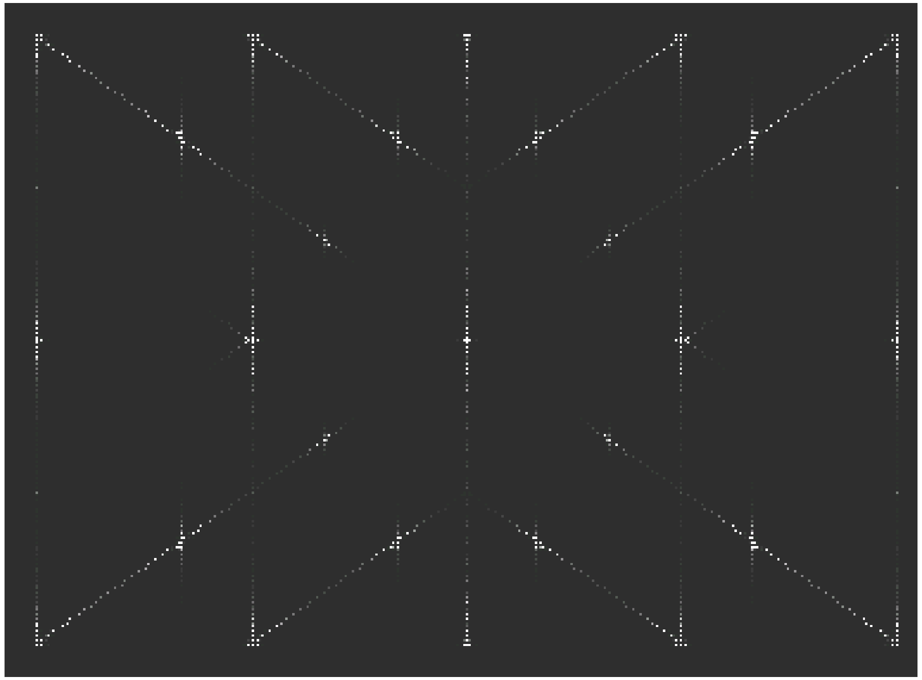
$n_{av} = 3x\omega(3) + 5x\omega(5) + 7x\omega(7) \ldots\ldots$

$|\Delta| = 1\text{-}3/n_{av}$

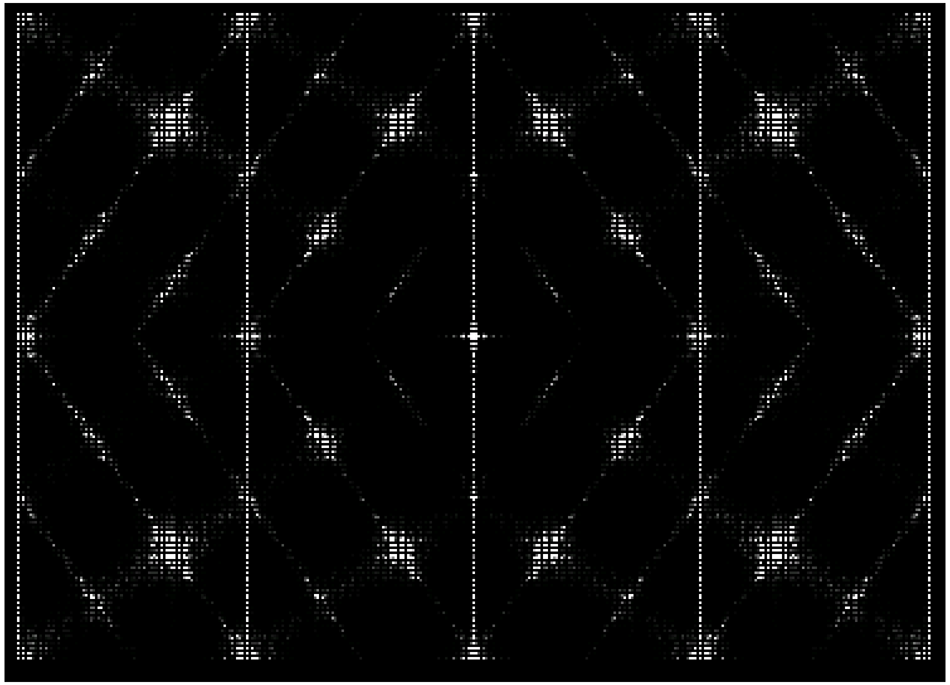
$$|\Delta| = \frac{R_{0001_R} - \frac{1}{3} R_{222_\beta}}{\frac{1}{6} R_{222_\beta}}$$



Simulation Results



One variant 50% ω , 100 lattice



84 % W (111) perfect U

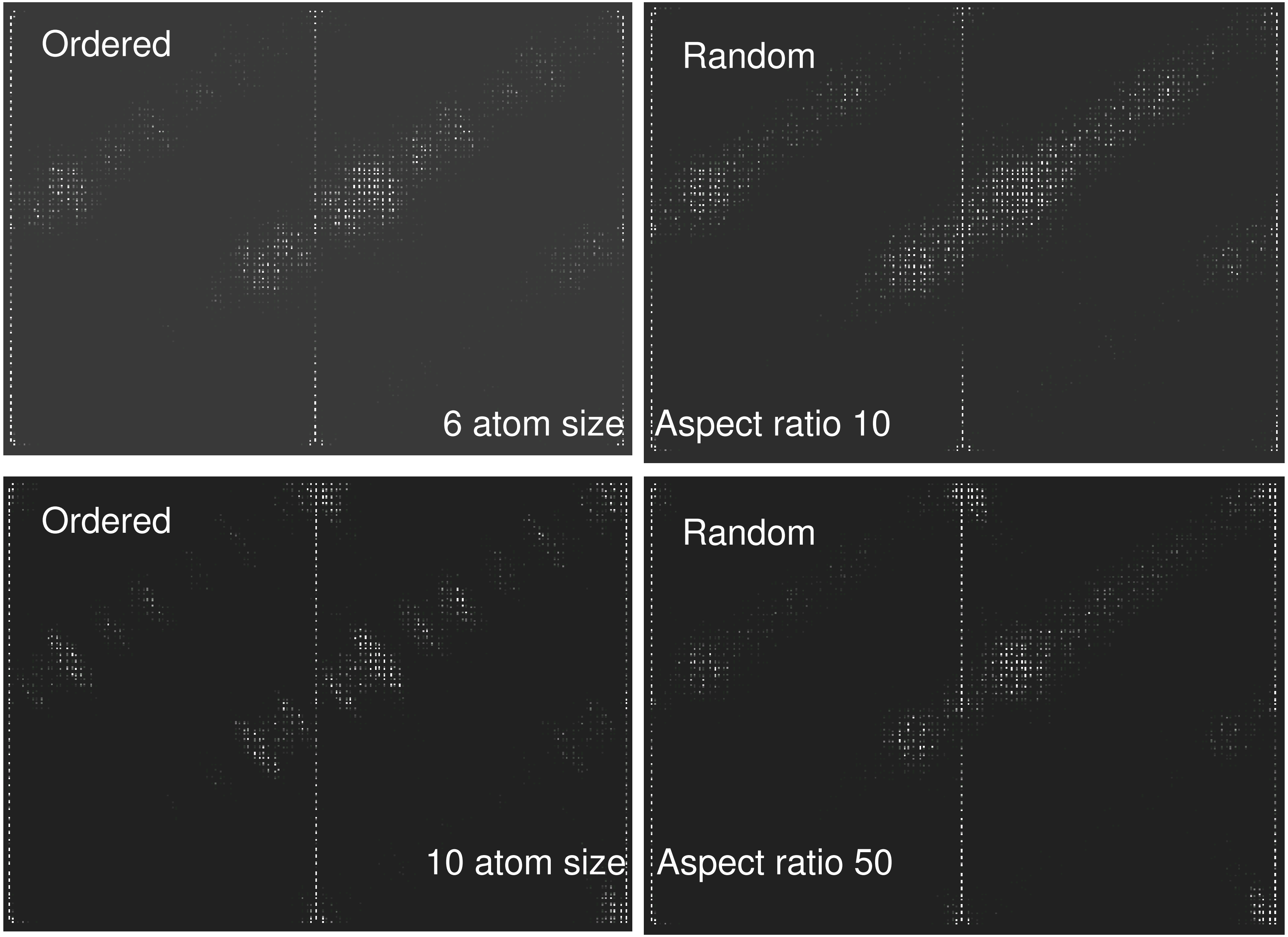
The streaking in the ω spots as observed in the SADs is reproduced by the present model by having rods nucleated along <111>

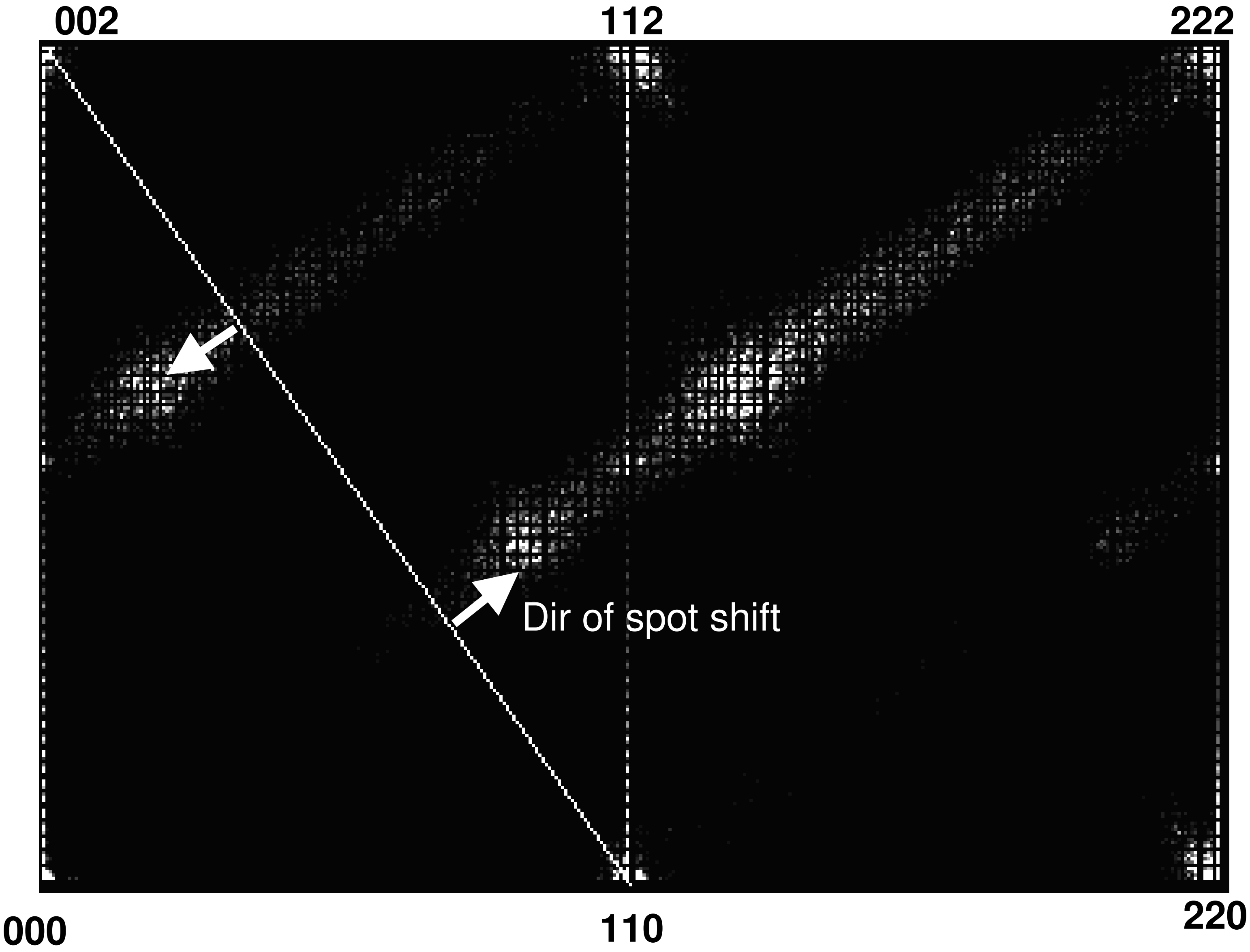
However the shift in the ω spots required the introduction of non ideal ω structural units like ω5 and ω7.

Earlier models (Sinkler and Luzzi) propsed that the shift in the ω spots is due to random sequence of different structural units of ω ie ω3,ω5,ω7.

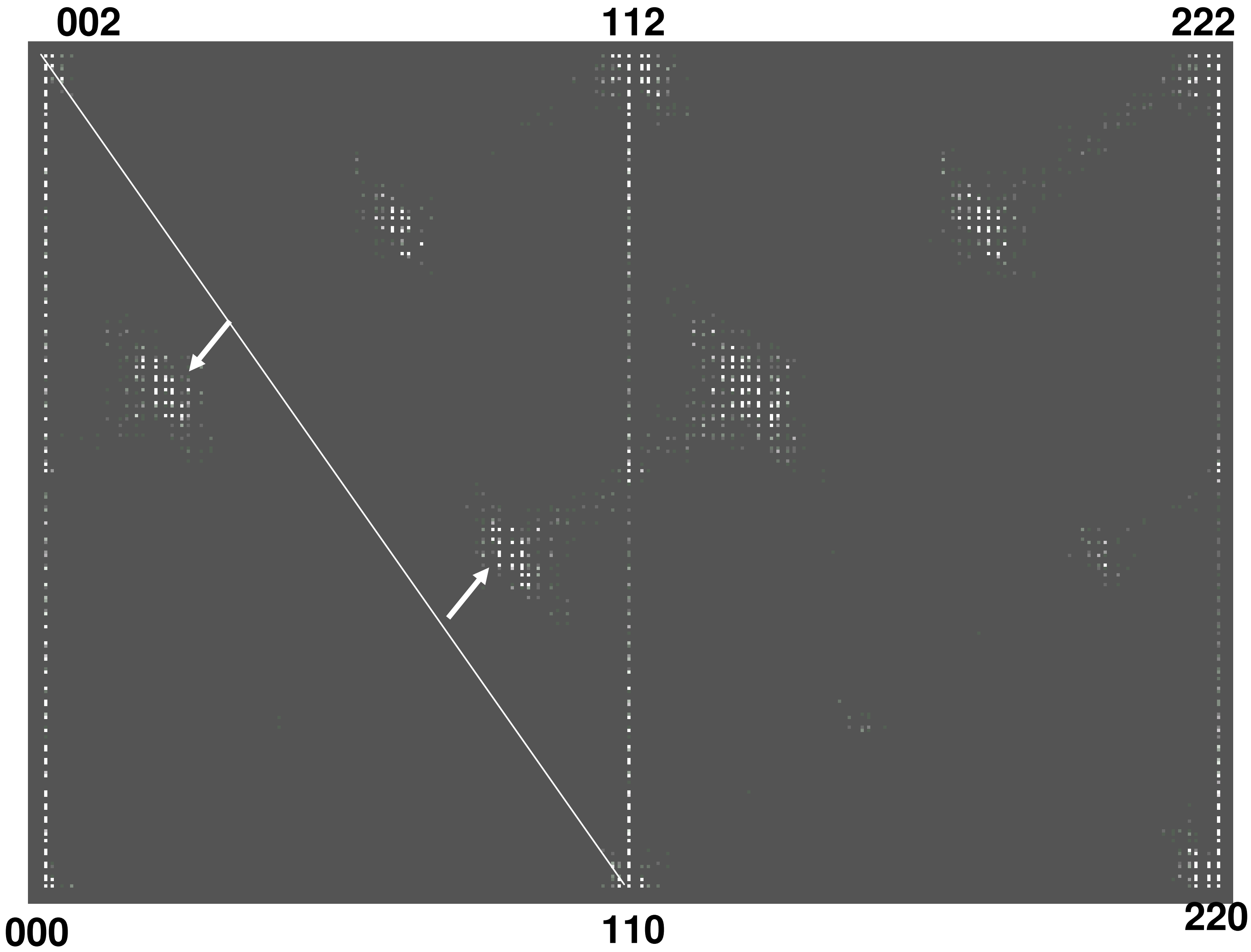
However when we simulated the diffraction pattern by nucleating ω rods in which each long road of ω composed of randomly sequenced ω3, ω5,and ω7 it failed to produce the observed streaking though it reproduced the shift in the spots.

It can be explained by the fact the streaking occurs as a result of a particular structural unit repeating itself in one direction. However if ω rod contains random sequence of ω3,ω5,ω7 it can't produce a single structural unit which is repetitive to cause the streaking.


Ordered
6 atom size
Random
Aspect ratio 10
Ordered
10 atom size
Random
Aspect ratio 50

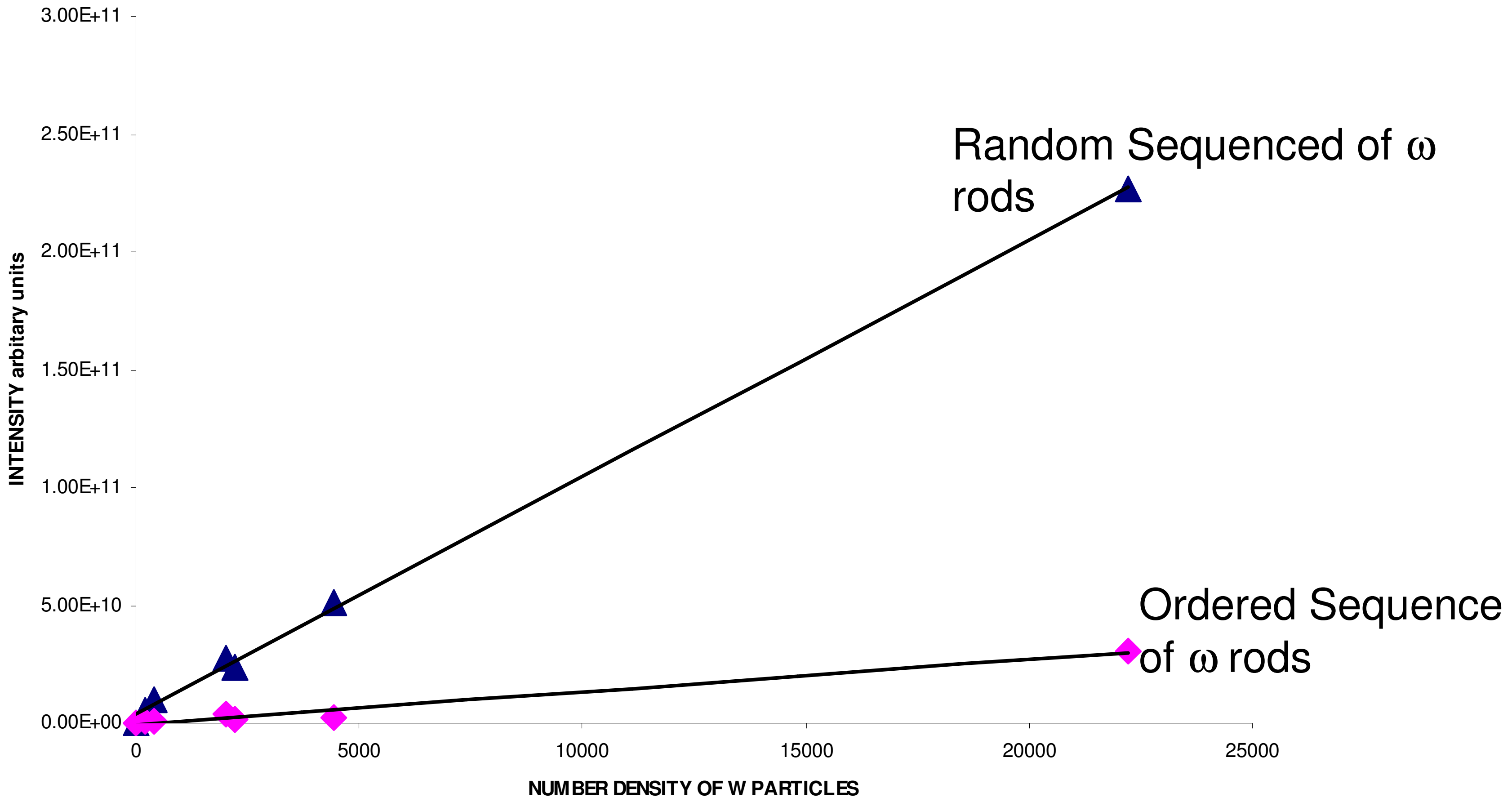


ω3, ω5, ω7 are randomly placed in ω rods.
Spot shifting can be observed. But streaking is not in the required direction, in spite of the fact that ω rods were generated along <111>
Also significantly high intensity at 111 is observed. It could be due to defect generated by presence of ω rods.



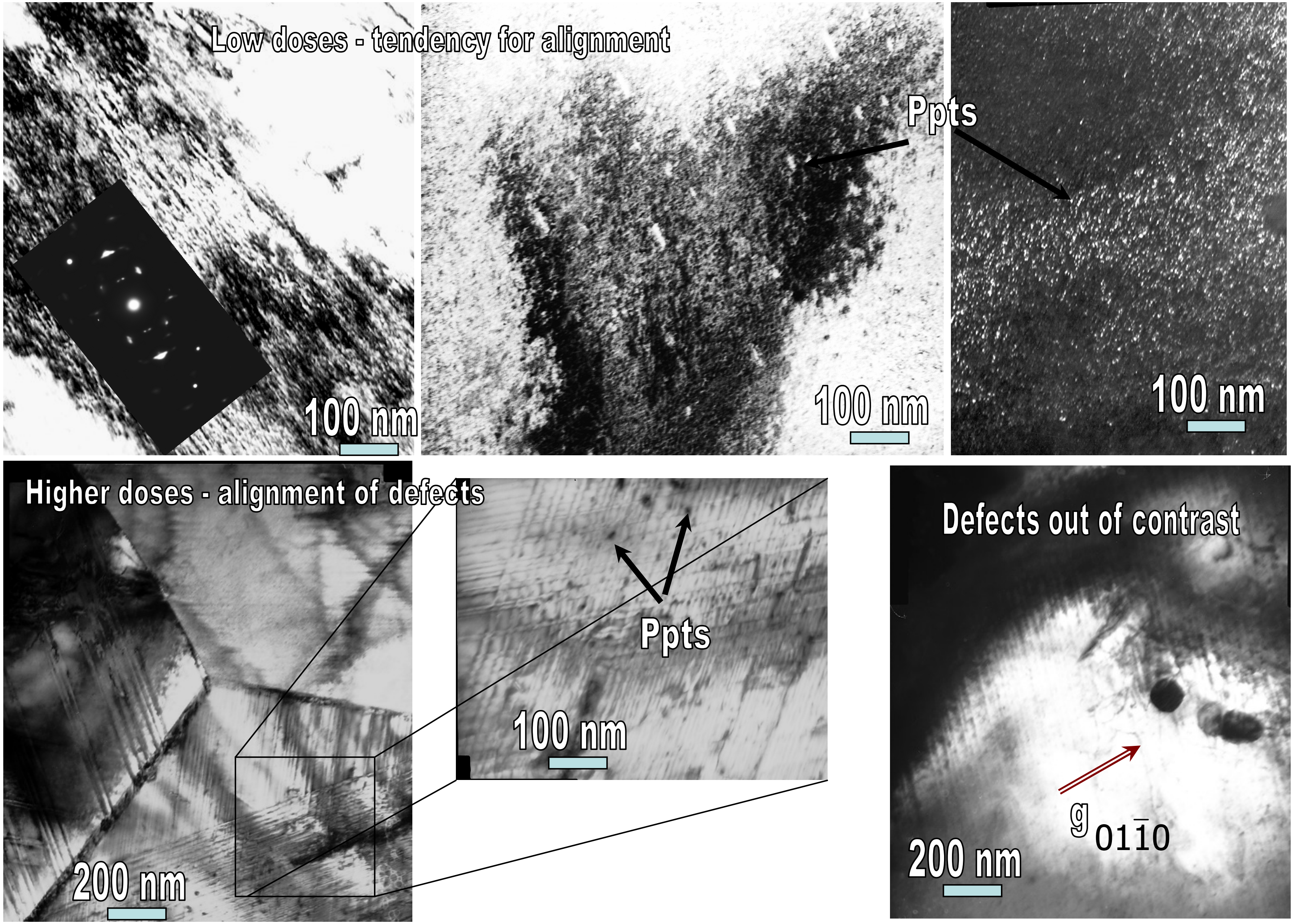
Here both streaking and spot shifting are produced. ω rods are generated by having ω3,ω5 and ω7 in sequence. Also the intensity of the 111 spot is reduced for the same ω number density as the previous picture.

Intensity of {111} reflection VS Number Density Of ω rods

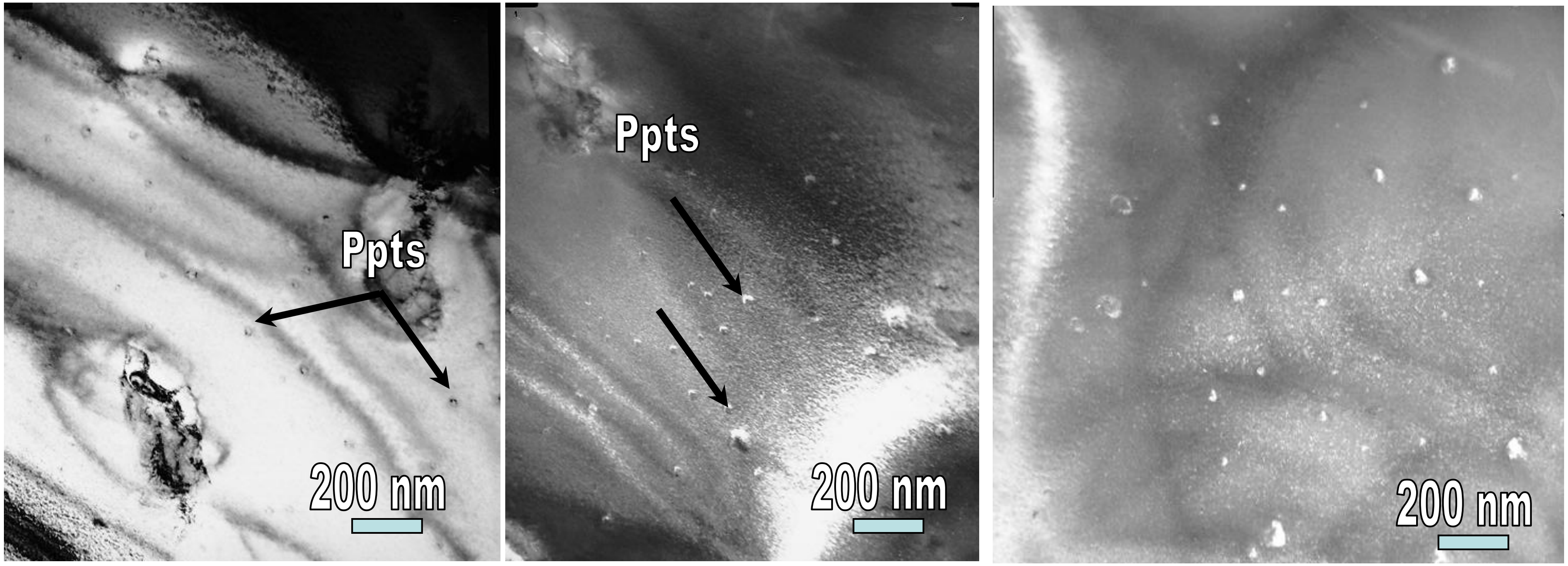


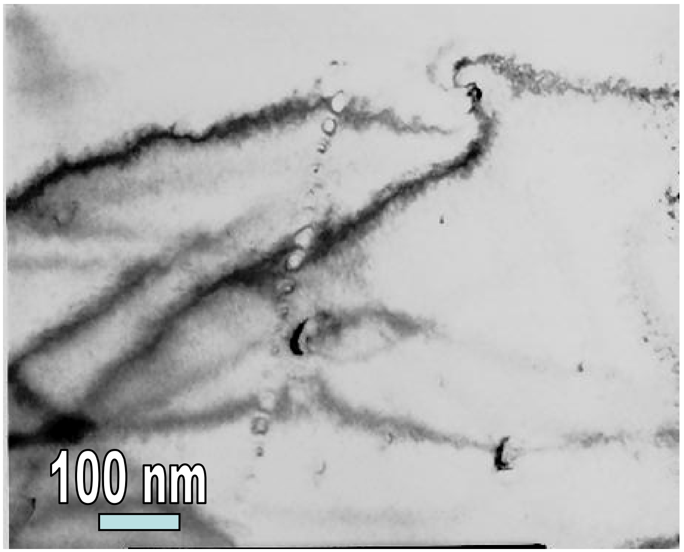
It is clear that 111 intensity also depends on ω number density and also on the nature of sequence of the ω3,ω5,and ω7

Irradiated Microstructures-formation of defects in Zircaloy-2



Irradiated Microstructures-formation of second phase in Zircaloy-2





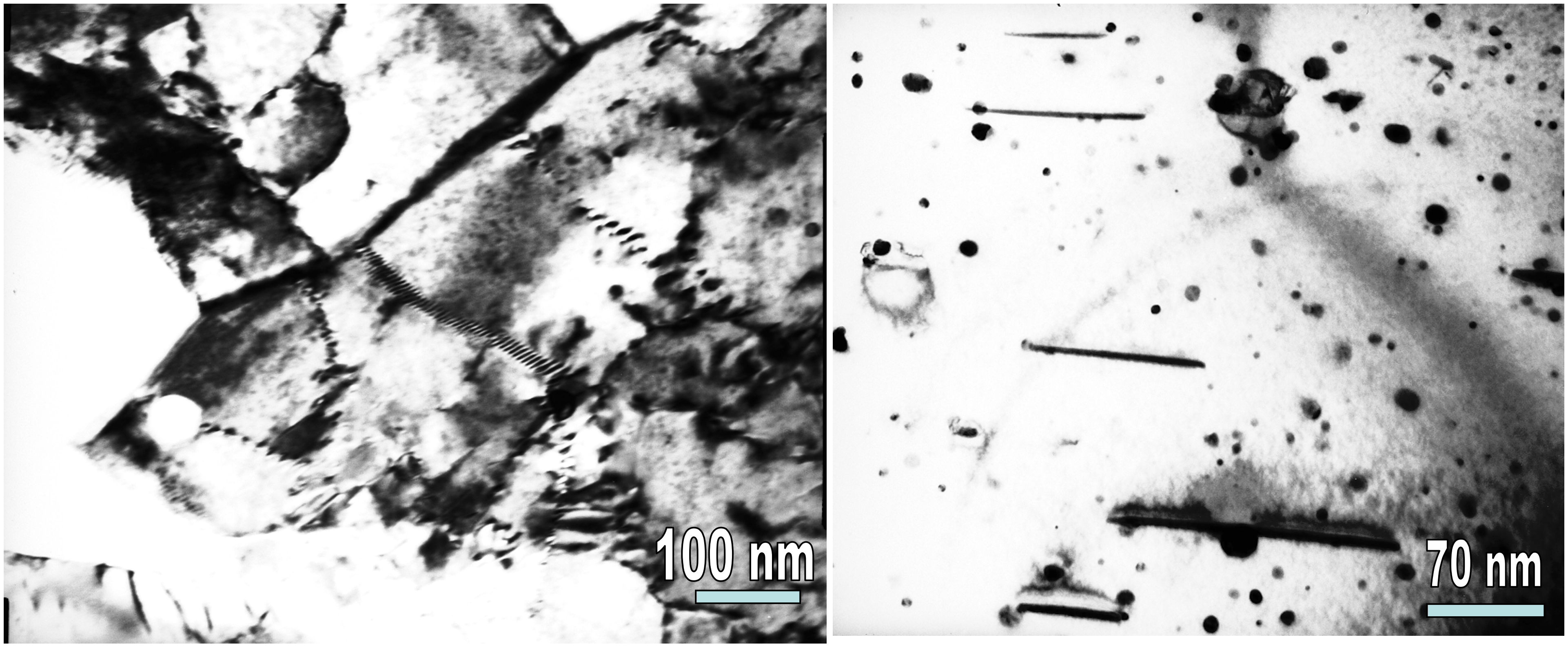
Formation of a phase was also noticed

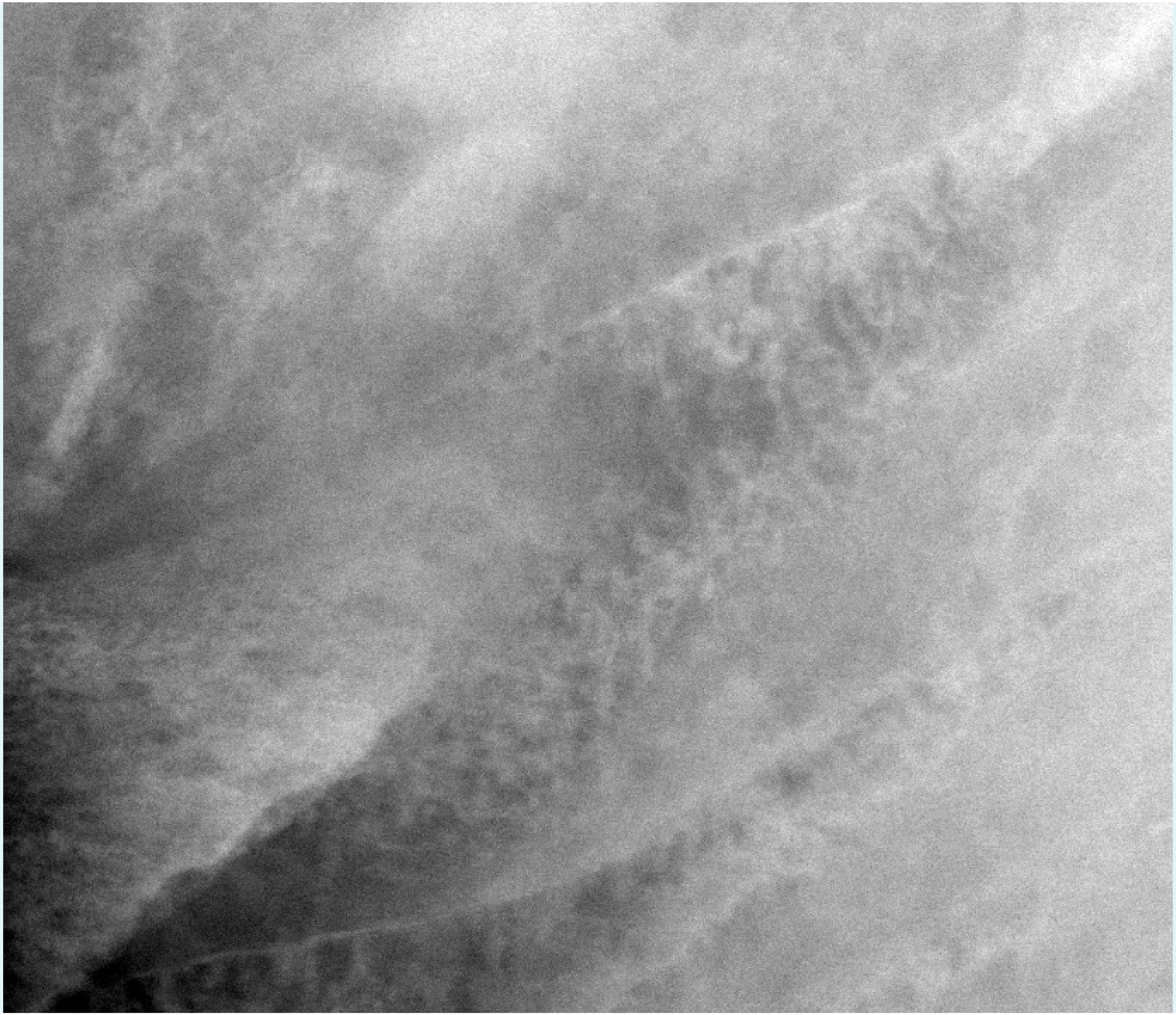
Size of the phase was less than 20 nm.

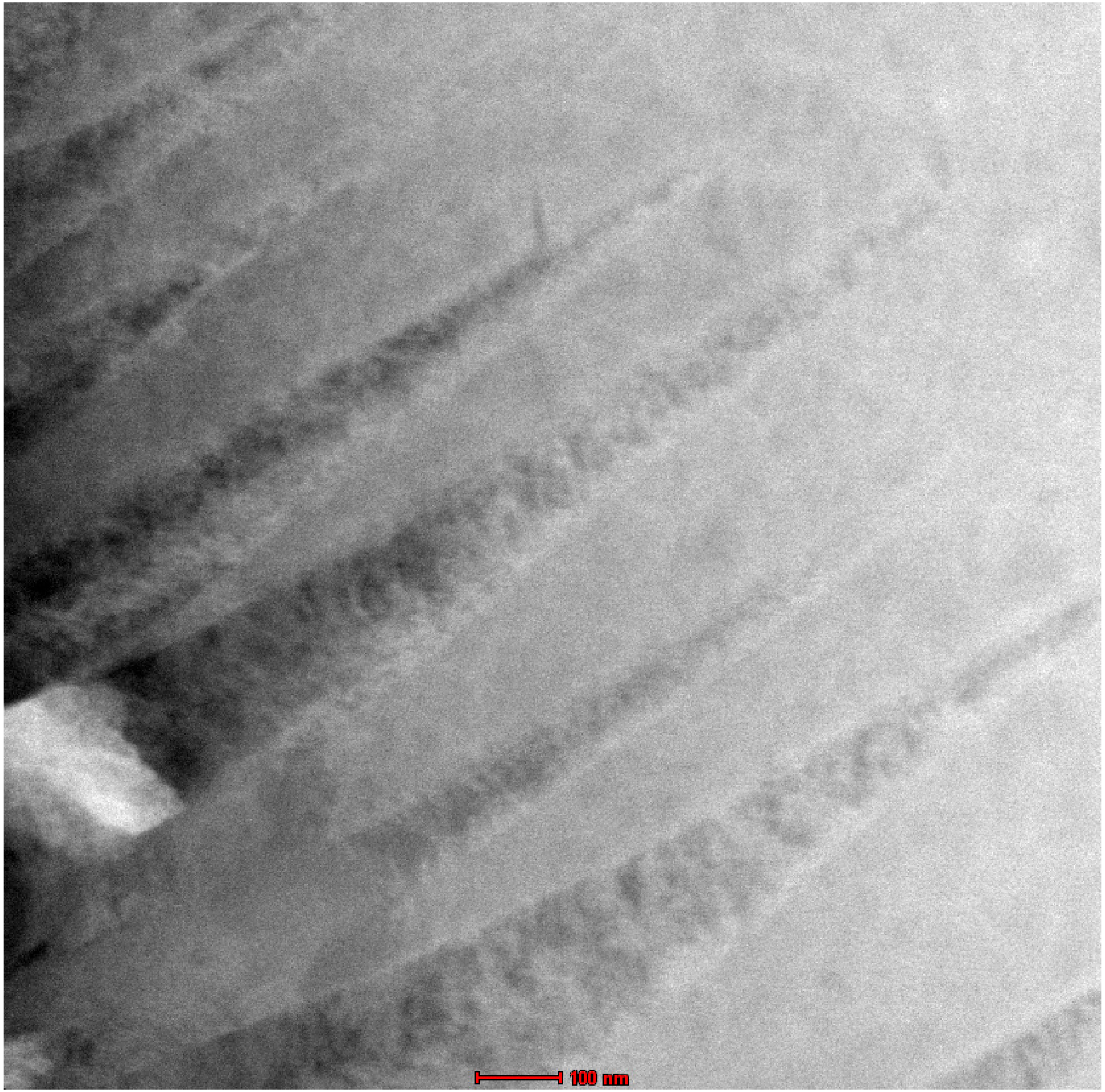
Distribution of the phase was random and uniform.

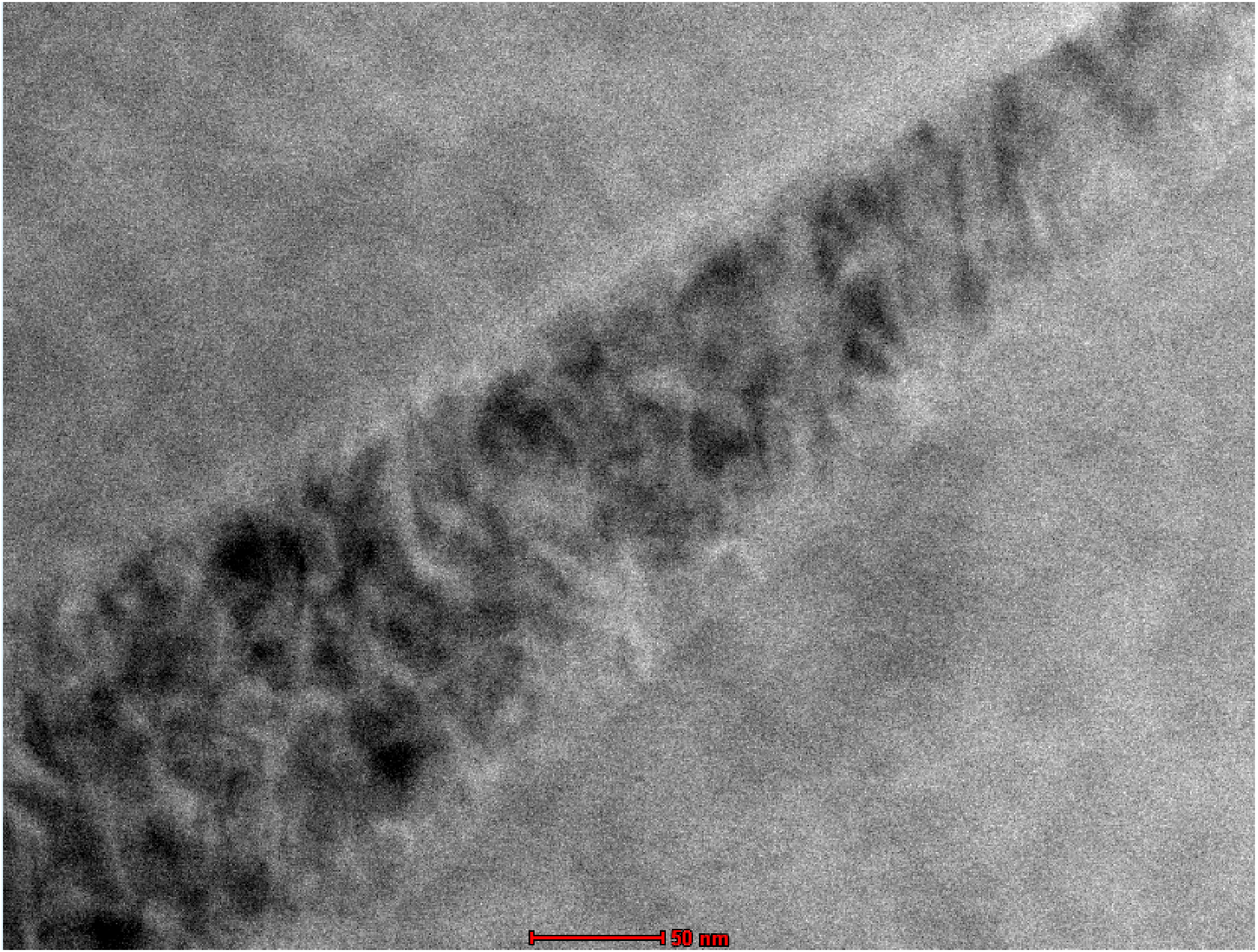
Some places alignment was observed.

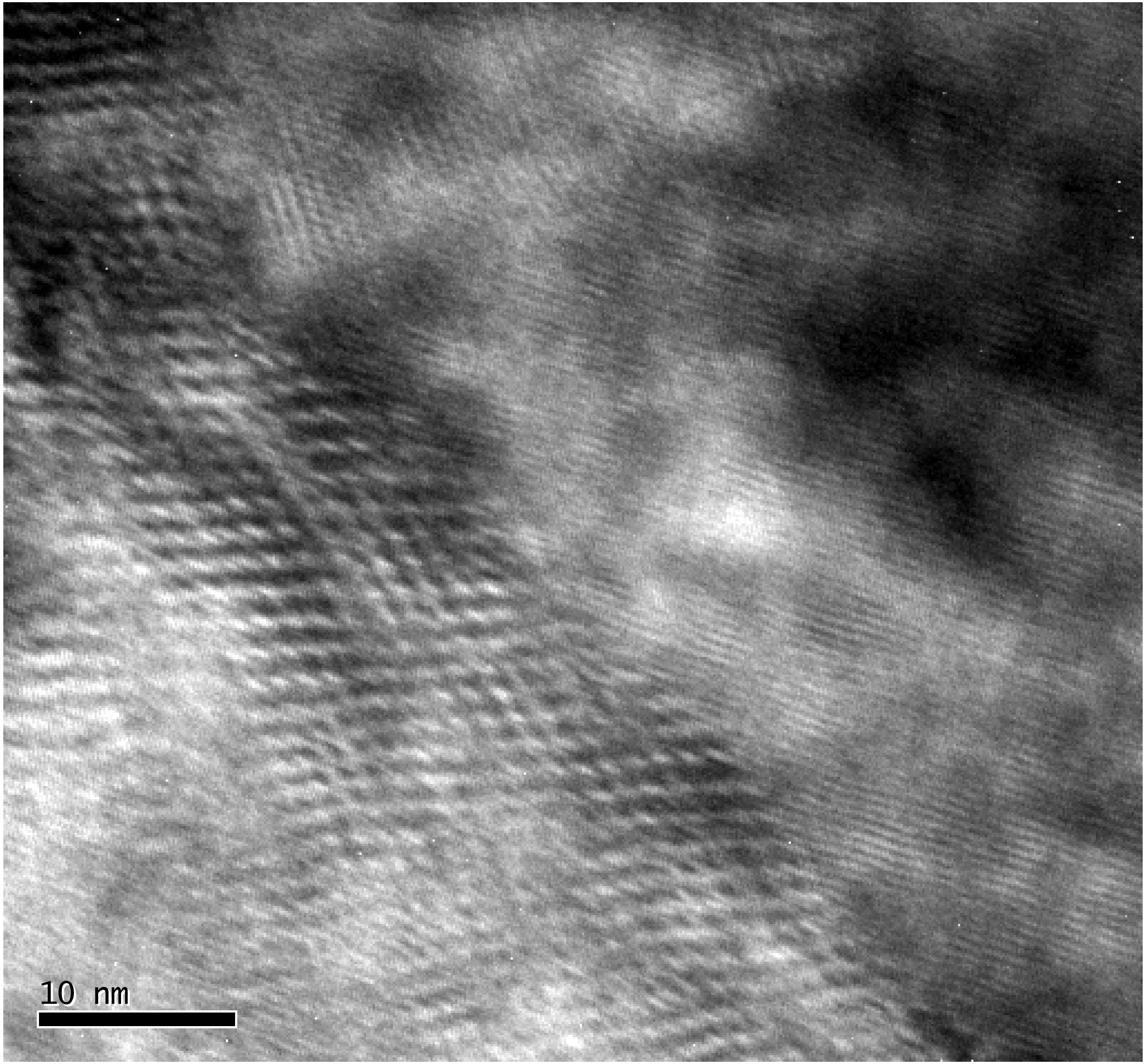
Zr-1Nb irradiation by oxygen ions

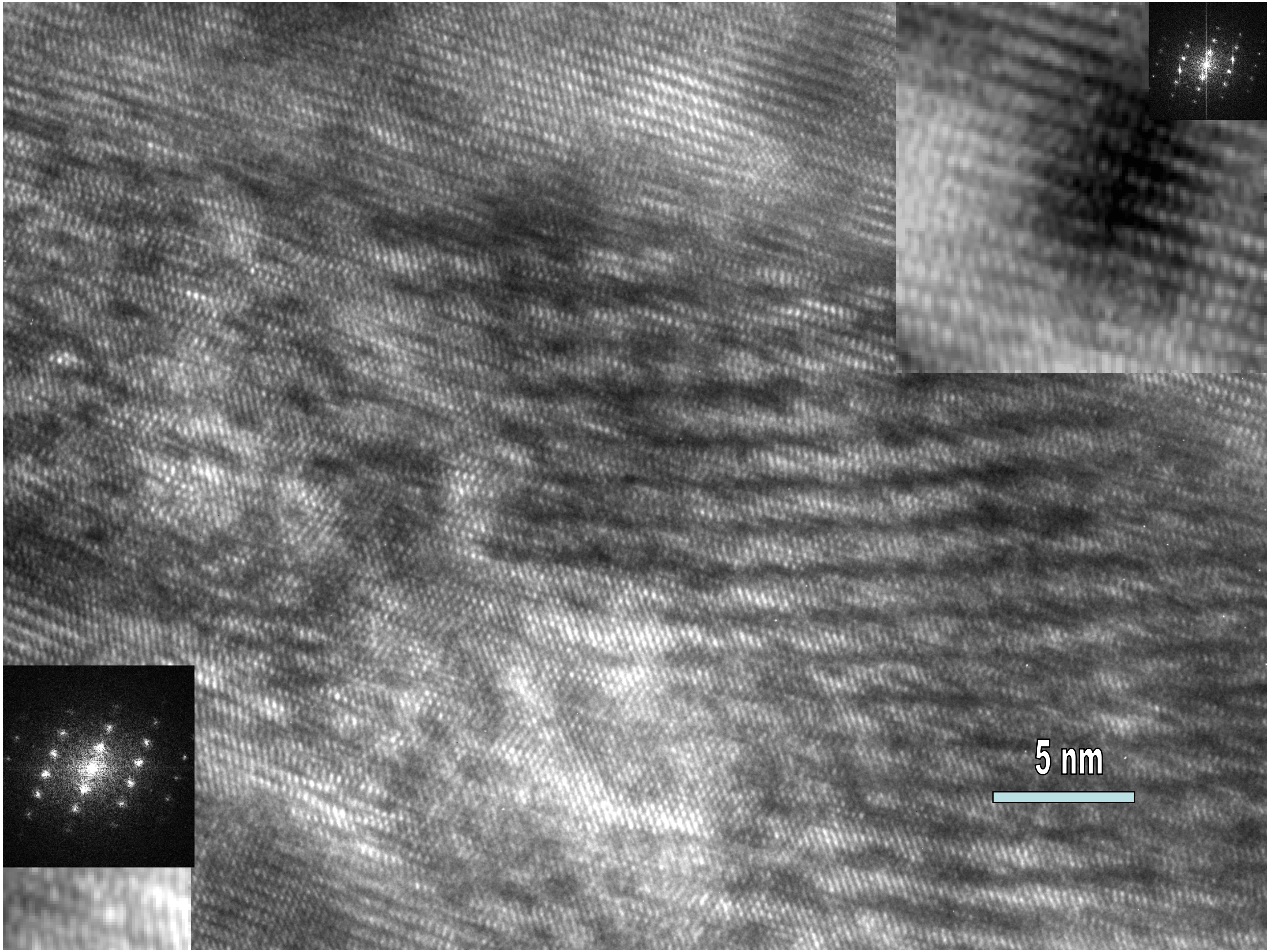


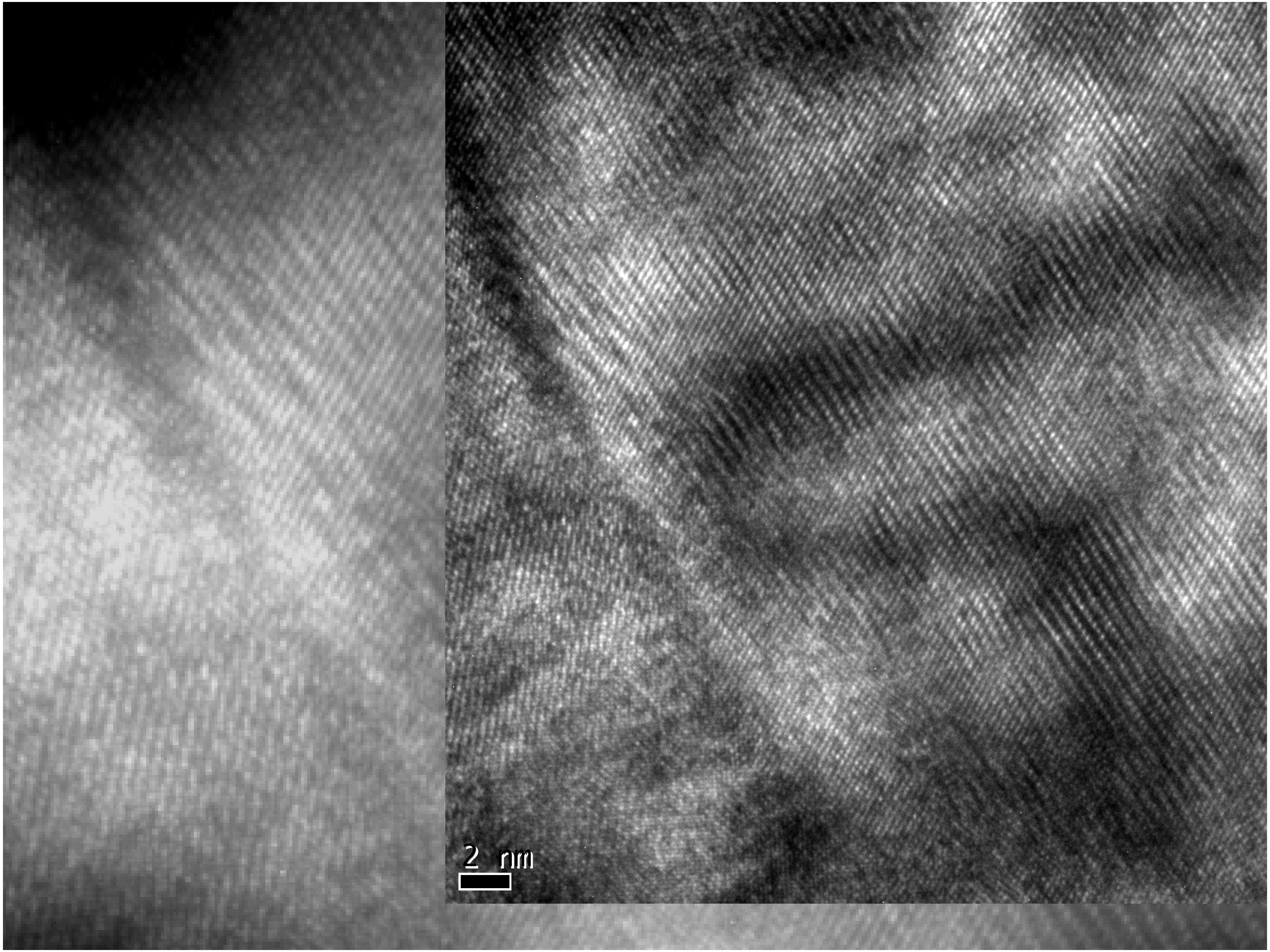



100 nm

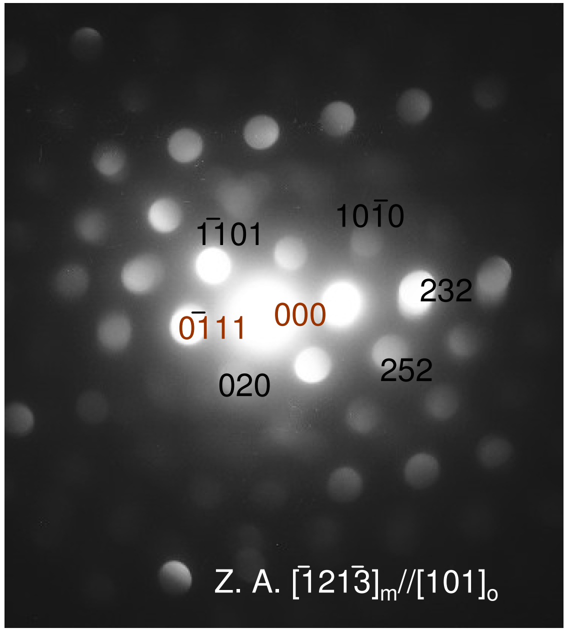

50 nm

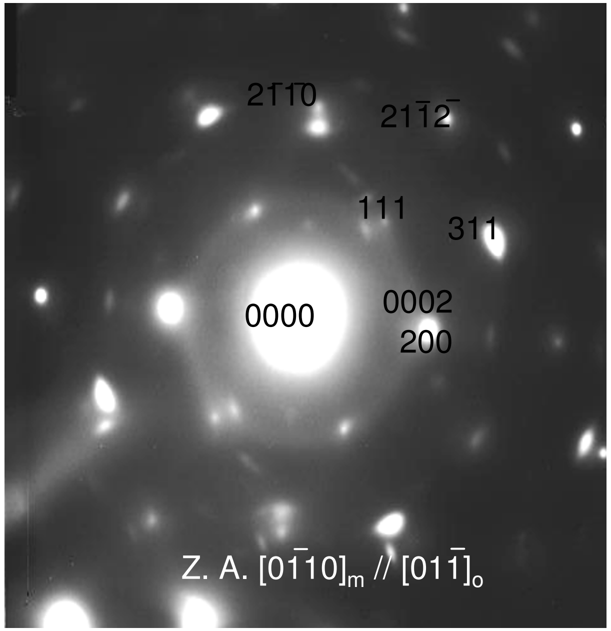

10 nm

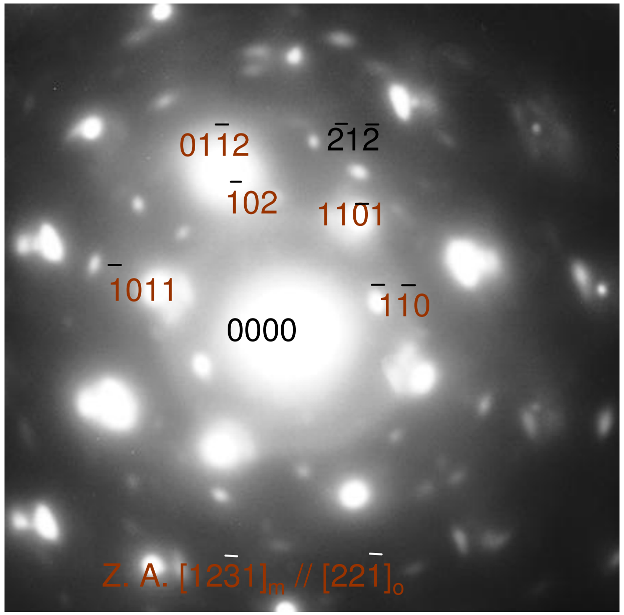

5 nm


2 nm

Composite diffraction patterns



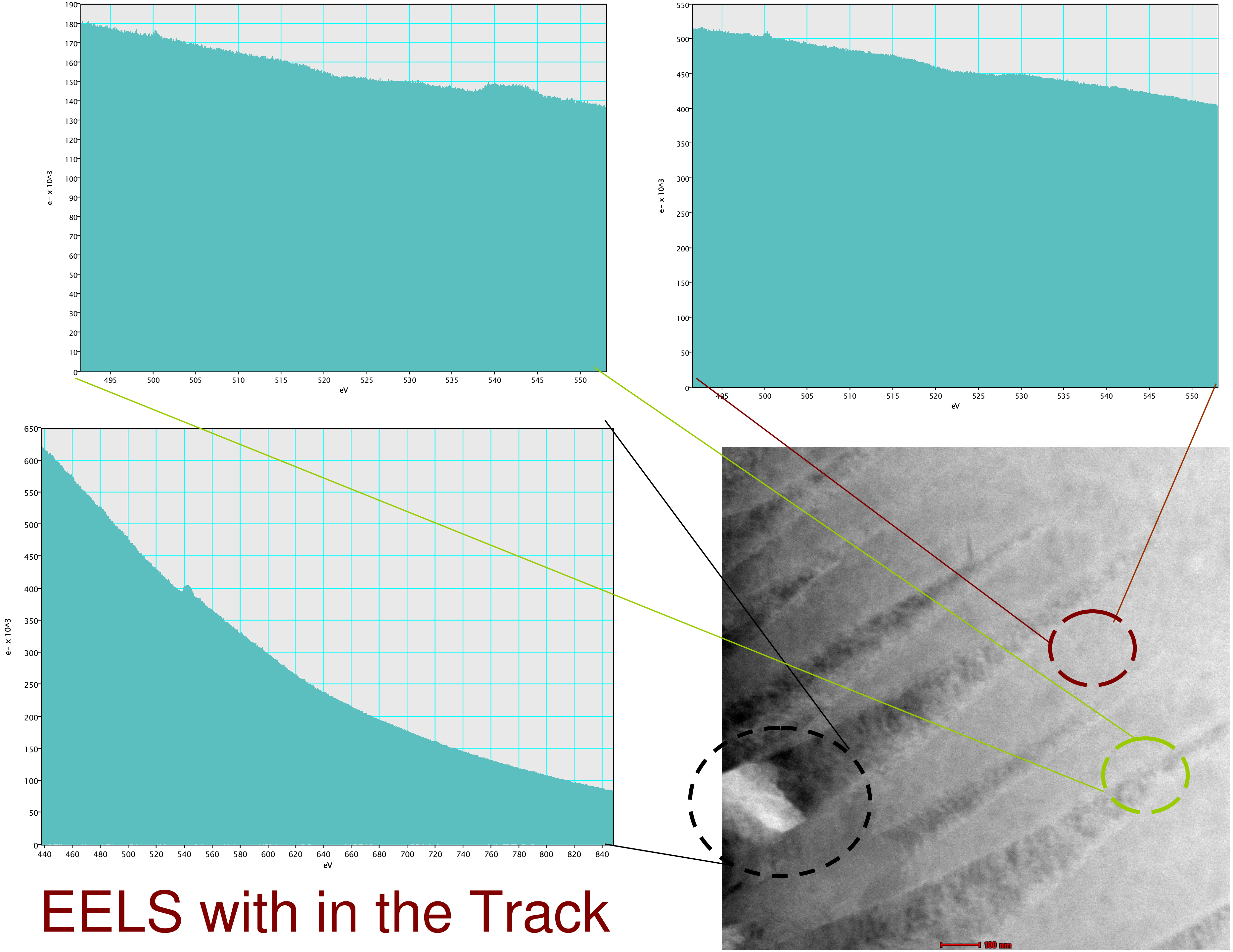




Patterns showed that

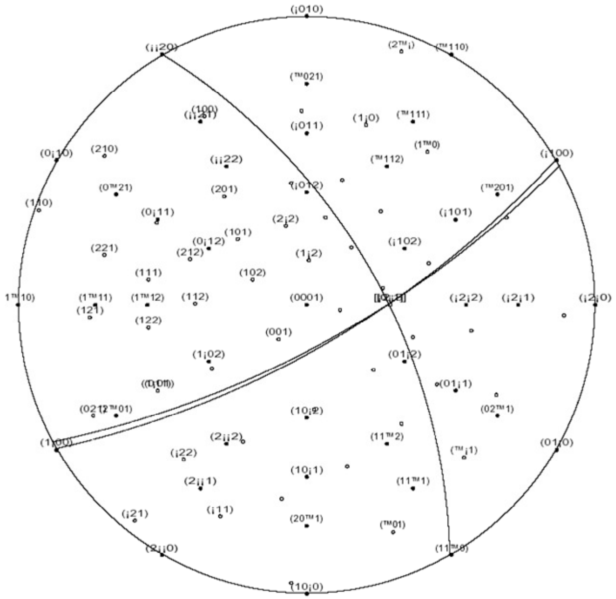
The phase formed during irradiation was the ZrO_2 phase.

Orientation relationship exists between the phases.



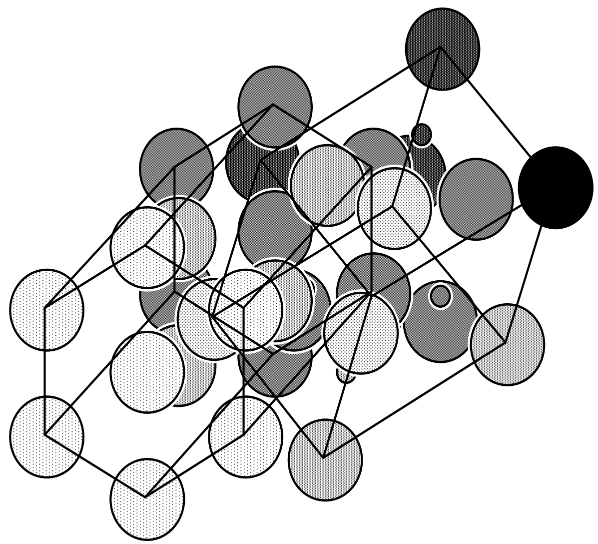
EELS with in the Track

Lattice correspondence between α and ZrO_2



Stereographic projection shows orientation relationship $(\bar{1}\bar{1}21)$ // (100) and $[\bar{1}2\bar{1}3]$ // $[0\bar{1}1]$

Based on OR- Lattice correspondence shows



Accumulation of Zr atoms at the (0001) plane

Possible Mechanism

Step one:

Irradiation induced point defects accumulate at Basal plane of the matrix a phase. Causing planar arrangement of defects.

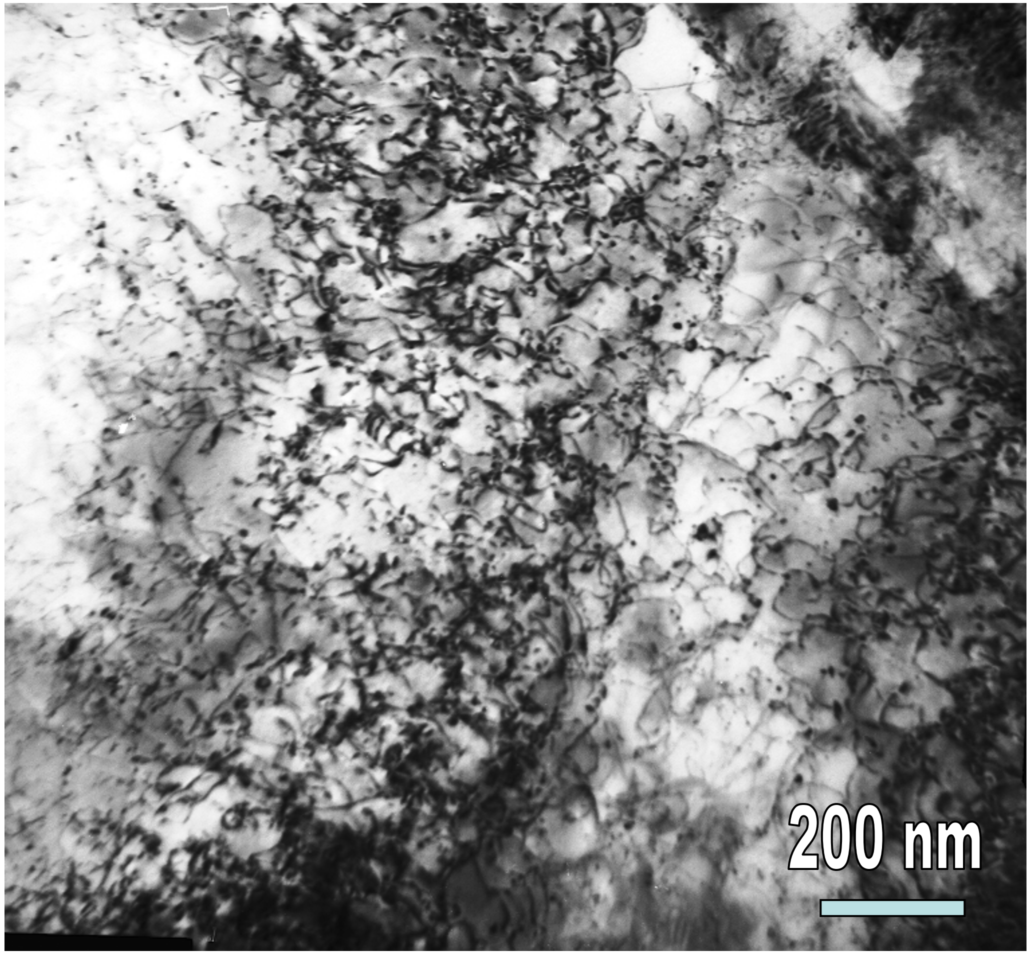
Step two:

Oxygen ions also get trapped into the matrix phase.

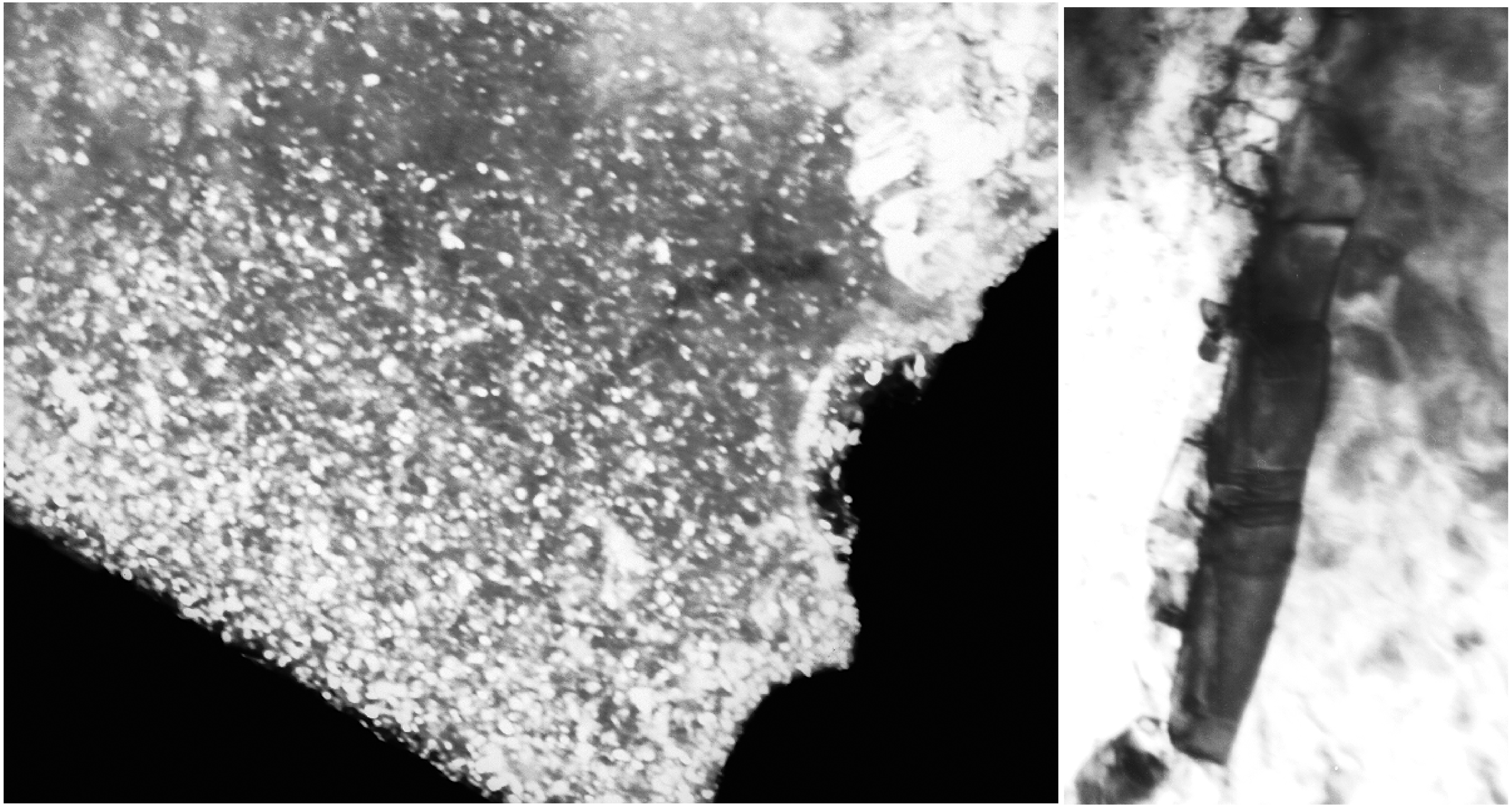
Step three:

Local strain and availability of the atoms precipitate the ZrO_2 phase.

Zircaloy-2 proton beam irradiated



Zircaloy-2 proton irradiation 8MeV



Summary

Effect of irradiation by ions has been studied in various zirconium based alloys.

Different defects and phase formed by irradiation have been identified. In case of zircaloy-2 samples majority of the defects were in form of the deformation bands. Microdiffractions and energy loss spectra indicated the formation of the Zr-O phase.

Upon electron irradiation, the β phase showed tendency for the formation of the ω phase. Simulations carried out showed that such difference could be due to possible arrangements of subunits in both the phases.

Proton irradiation showed the formation of β phase in α-Zr.